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Administrative comments -
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OFFICE OF CONTRACT ADMINISTRATION

NOTICE OF PROJECT CLOSEOUT

Closeout Notice Date 02/01/90
Original Closeout Started ********

Project No. E-25-M34 Center No. R6445-0A0

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NOTE: Final Questionnaire sent to PDPI.

A STUDY OF ELECTROCHEMICAL PARTITIONING IN NUCLEAR FUEL REPROCESSING

FINAL REPORT
PROJECT E25-M34

Project Director: A. Schneider

Submitted to

Martin Marietta Energy Systems, Inc. - ORNL
P.O. Box X
Oak Ridge, TN 37831

January, 1990

Work done under Subcontract No. 19X-SB386C
with Martin Marietta Energy Systems, Inc.

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All on a part-time basis.

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FIGURES

SUMMARY

A bank of eight 2-cm ANL type annular centrifugal contactors was tested hydraulically and its operability range was determined. Uranium (VI) extraction and stripping runs were made and the experimental results agreed well with those estimated with the SEPHIS MOD (IV) computer code. Rapid analytical techniques were developed for uranium (VI), uranium (IV), and free nitric acid determinations in aqueous and organic solutions.

An evaporator with separate electronic temperature and pressure controls was designed and constructed for waste stream clean-up and uranium recovery. A platinum/titanium electroreduction cell was designed and built. The cell was tested and the results were used to develop a mathematical model for U (VI) reduction (Program ELECTRO). The contactor was hydraulically tested a second time, with the electrocell placed in-line, between stages 1 and 2 (stage 1 being the aqueous entrance).

The SEPHIS MOD (V GT) program was developed to allow for modeling the simultaneous reduction of U (VI) and Pu(IV) as well as multicomponent extraction. The results estimated with SEPHIS (V-GT) agreed well with the experimental results when an argon blanket was used to prevent U(IV) reoxidation.

The research program is shown schematically in Figure i.1.

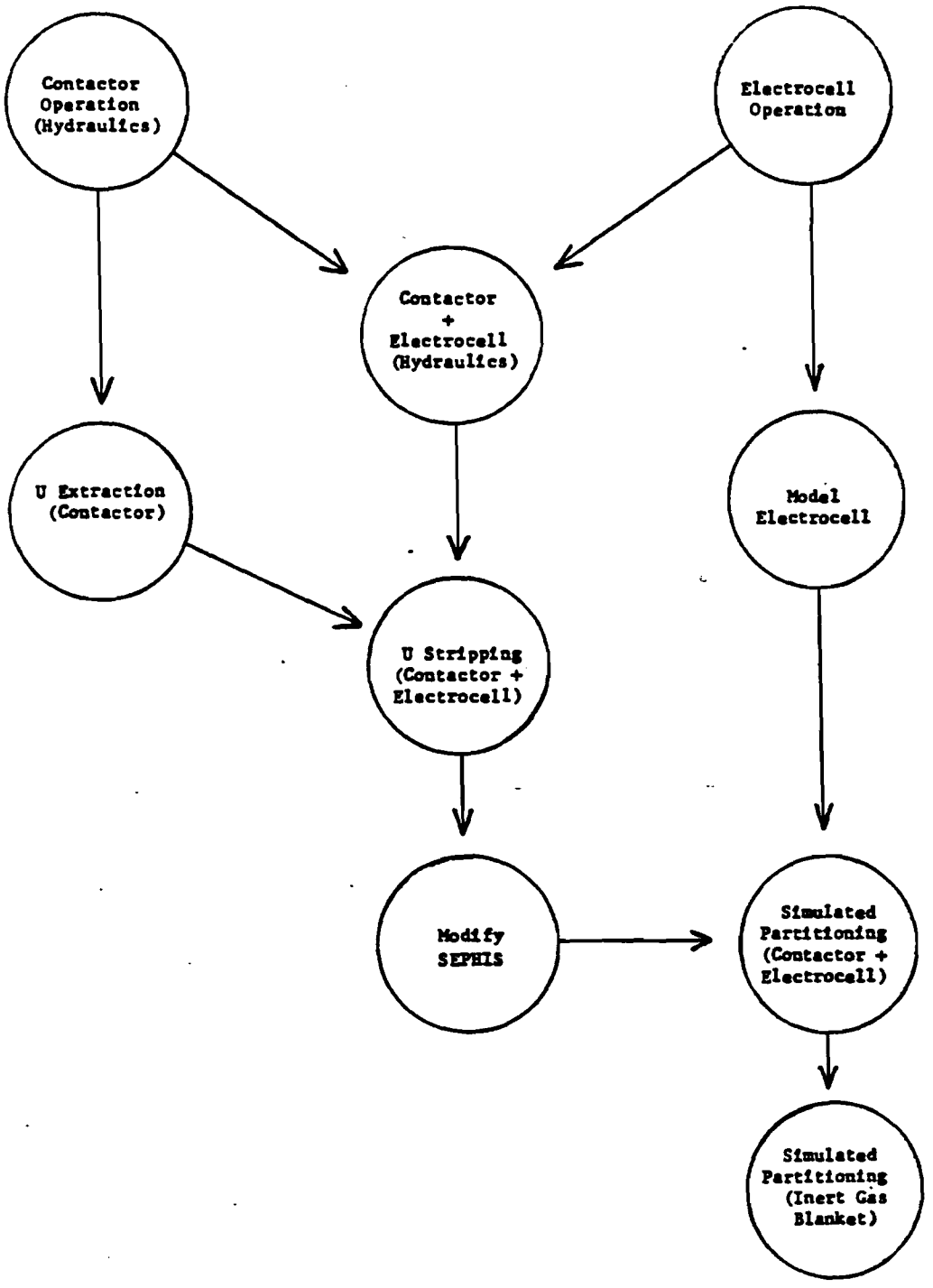


Figure i.1 Outline of Research Program

1. INTRODUCTION

The reprocessing of irradiated nuclear fuel, from LWRs and LMFBRs, to separate reusable uranium and plutonium from transuranic elements and fission products, has been accomplished at reprocessing facilities around the world since the 1950's. The most widely adopted method for fuel reprocessing has been the Purex process which employs solvent extraction with TBP in a hydrocarbon diluent as the extractant. Since the process was originally developed, there have been many attempts to improve the technology and the economics. The uranium/plutonium separation step has received much attention because of some undesirable features of the original Purex process.

The uranium/plutonium separation involves the reduction of plutonium (IV) to plutonium (III) which has been accomplished by the addition of the chemical reductant/stabilizer ferrous sulfamate. Large quantities of ferrous sulfamate are required, however. This leads to an increase in extraneous chemicals in the initial process stream and to the formation of significant quantities of waste salts, such as ferric sulfate [17][22]. These salts complicate subsequent waste processing and increase corrosion of the stainless steel process equipment and liquid waste storage tanks [24][28].

Many studies have been made to minimize or avoid the production of ferric sulfate and other salts. One of the more promising concepts has been the replacement of chemical reductants (ferrous sulfamate, HAN, etc.) with the

direct electrochemical reduction of Pu(IV) and U(VI). The U(IV) thus formed reduces Pu(IV) in the process stream [29]. This technique introduces no extraneous solids, since unreacted U(IV) reoxidizes to U(VI) and the hydrazine stabilizer which is normally used can be subsequently destroyed by chemical or electrochemical means.

The high plutonium content of breeder reactor fuel may favor the use of the electrochemical method. This is due to the fact that the reductant required to reduce the greater amounts of plutonium present leads to an increase in the mass of salts formed [25][27].

In the processing of spent fuel it is important to use solvent extraction contactors in which the time of contact between the highly radioactive aqueous stream and the organic solvent is minimized, to prevent radiation damage to the TBP/diluent extractant.

Multistage centrifugal contactors have many advantages over other extractor types [17][20][5]. There also appear to be some advantages to the combining of centrifugal contactors with electrochemical cells for plutonium/uranium partitioning.

Computer codes developed to model solvent extraction processes are valuable tools in process design and analysis. They permit the optimization of process conditions and provide an indication of the distribution of various species throughout the cascade. This is important in criticality safety considerations and in the investigation of transient effects. The computer

program SEPHIS, developed at the Oak Ridge National Laboratory to model counter-current solvent extraction cascades, has been found to be particularly useful for modeling the Purex process in mixer-settler type extractors [19][37]. The objectives of this study were the testing of a reduction cell operating in tandem with a bank of centrifugal contactors, the modification of SEPHIS for modeling such a system, and the utilization of computer modeling in assessing the adequacy of electrochemical partitioning in the reprocessing of breeder reactor fuel.

2. BACKGROUND

2.1. The Purex Process

The Purex Process consists of a series of solvent extraction cycles which allow the separation of U and Pu from fission products and other impurities and the partitioning of Pu and U from each other. In solvent extraction, a liquid solvent is added to a solution containing a solute. During mixing of the two immiscible liquids, one liquid becomes temporarily dispersed in the other "continuous" phase. The solute distributes between the two liquid phases until equilibrium is reached. The resulting solvent solution, now containing some of the solute, is termed the extract and the solute solution, now containing less of the solute, is called the raffinate [1][2].

The distribution coefficient is a measure of the solute's relative affinity for the two phases and is defined by the following equation [2][1].

$$D_A = Y_A/X_A$$

where, D_A = The distribution coefficient at equilibrium.

Y_A = The solute's concentration in the extract at equilibrium.

X_A = The solute's concentration in the raffinate at equilibrium.

The distribution coefficient is not a constant but varies with the concentration of the solute, the temperature, and the presence of other species in the solutions. The rate of extraction of the solute generally increases with temperature [2]. In certain systems, such as the TBP-U-HNO_3 system, the distribution coefficient varies with the acidity of the solute solution and with the valence state of the solute to be extracted [1][3]. The first property can be used to increase or decrease the extraction of a particular element. The second can be exploited to preferentially transfer an element from one phase to another by changing its valence.

In the Purex process, uranium and plutonium are transferred from an aqueous, nitric acid solution into an organic liquid (normally 30% TBP, by volume, dissolved in a hydrocarbon diluent to reduce the viscosity) [1][3]. Transfer occurs because uranium and plutonium form complexes with TBP and nitrate ions that are soluble in the organic diluent in accordance with the following equations [15][3]:

$$UO_2^{2+}\ (aq) + 2NO_3^-\ (aq) + 2TBP\ (org) \rightleftarrows UO_2\ (NO_3)_2 \cdot 2TBP\ (org)$$

$$Pu^{4+}\ (aq) + 4NO_3^-\ (aq) + 2TBP\ (org) \rightleftarrows Pu\ (NO_3)_4 \cdot 2TBP\ (org)$$

These equations show that the equilibrium can be affected by the nitrate (NO_3^-) concentration. Thus, to transfer UO_2^{2+} or Pu^{4+} from the aqueous to the organic phase, the concentration of nitrate ions must be high. This is accomplished by increasing the concentration of nitric acid (generally an initial nitric acid concentration of about 2.5 N or 3.0 N is enough to maximize the distribution coefficient [7]) or of other "salting agents" such as aluminum nitrate. Conversely, contacting the loaded solvent (the extract) with pure water, decreases the nitrate ion concentration and causes the TBP-metal complexes to decompose. The uranium and/or plutonium transfer back into the aqueous phase. This step is defined as back-extraction or stripping.

To separate the uranium from the plutonium, use is made of the fact that, while U(IV), U(VI), and Pu(IV) form strong organic complexes with TBP, Pu(III) forms a relatively weak TBP complex [15][24]. This means that plutonium in its reduced state (Pu(III)) can be back-extracted into a nitric acid solution, while the uranium remains complexed in the organic phase, as long as the acidity of the aqueous phase is sufficiently high [7][3]. This step is defined as partitioning.

2.2. Solvent Extraction Equipment

Different types of equipment have been used for liquid-liquid extraction. Solvent extraction equipment (generally referred to as contactors) can be classified into two main categories: equipment with discrete stages, where the

two liquids are mixed, allowed to settle and the immiscible phases are separated, and differential type contactors. The first type consists of single discrete stages, each providing mixing, settling, and separation, which are connected to form a cascade. This type of equipment is referred to as mixer-settlers. In centrifugal contactors centrifugal force is used to accelerate the settling.

The second type of equipment, which includes pulse columns, provides for continuous, or differential, contact. Unlike mixer-settlers, this equipment allows the immiscible liquids to flow countercurrently, without periodic settling and physical separation from each other. Continuous contactors can be built to contain the equivalent of many stages, without the intermediate removal of the phases.

In pulse columns, the aqueous phase, which is usually heavier than the organic phase, is introduced at the top of the column and the organic solvent at the bottom. Perforated plates separate the two phases when the liquid is at rest. One liquid is periodically pulsed to force the organic to the top and the aqueous phase to the bottom of the column, where they are, respectively, drawn off.

Initially, only gravity type mixer-settler units were used, but later, mixer-settlers using centrifugal force for phase separation (centrifugal contactors) were developed at the Savannah River Laboratory. Advantages of the design include a high extraction efficiency, high throughput (i.e., short residence time),

rapid start up and shutdown, and small stage volumes. Centrifugal contactors have been used at the Savannah River Plant since the mid-1960's [16].

In the late 1960's, a modified version of the SRL centrifugal contactor was developed at the Argonne National Laboratory. The ANL contactor, also known as an annular centrifugal contactor, was simpler, fabrication costs were lower and it was more amenable to remote maintenance. The improved ANL design also retained all of the advantages of the SRL design [5][16].

The annular centrifugal contactor is especially attractive for use in reprocessing breeder reactor fuels, which contain higher concentrations of plutonium and fission products than LWR fuel. This is because the short residence time of the liquids in the contactor reduces the radiation exposure of the organic solvent and, thus, the degree of TBP degradation (see section 3.6.3), [20][17]. The small holdup volume and small characteristic diameter of each stage permits practical plant scale operations in units that are sized to be critically safe [17][5].

2.3. Plutonium Reduction Options

2.3.1. Chemical Reduction

Traditionally, the reduction of Pu(IV) to Pu(III) has been accomplished by the addition of ferrous sulfamate, with the ferrous ion (Fe(II) acting as the reducing agent and the sulfamate (NH_2SO_3) ions acting to stabilize the Pu(III)

and Fe(II) ions in the normally oxidizing nitric acid medium [22][28]. The reaction is as follows [3]:

$$Fe^{2+} + Pu^{4+} \longrightarrow Fe^{3+} + Pu^{3+}$$

The use of ferrous sulfamate has the disadvantage that the subsequent neutralization of the raffinate (high-level liquid waste) leads to the formation of insoluble iron hydroxides and sulfate salts [22]. This is undesirable, since the introduction of these extraneous salts adds considerably to the volume of the waste and complicates the eventual solidification. SRP reported that an almost forty-fold excess of ferrous sulfamate was often required to reduce the Pu(IV) present, thus making the ferrous sulfamate reagent the single largest source of waste solids from the process [7][22]. In addition, the extraneous salts were found to significantly accelerate the corrosion of stainless steel process equipment in the plant [24][28].

The disadvantage of using ferrous sulfamate is even more serious for reprocessing the spent fuel from LMFBR's, where the plutonium content is so high that a ten fold increase results in the amount of undesirable salts introduced to the process stream.

An alternative reductant, HAN (hydroxylamine nitrate) was developed to reduce the amount of solid waste in the process stream. SRP found that they could reduce the amount of ferrous sulfamate required, and thus the waste solids formed, by one third when using HAN [22]. Although HAN has the

advantage that it decomposes safely to gaseous nitrogen oxides and water upon oxidation [22], its Pu(IV) reduction rate is slow and decreases with increasing nitric acid concentration [24]. HAN has also been found to allow some plutonium, bound to TBP degradation products, to remain in the organic phase [6].

A further reduction in waste solids can be obtained by replacing the previous reductants with uranium (IV) [13][16]. The plutonium (IV) reduction occurs according to the following reaction [3][24].

$$2Pu^{4+} + U^{4+} \longrightarrow 2Pu^{3+} + U^{6+}$$

Hydrazine has been the stabilizer of choice, to prevent the reoxidation of Pu(III) and the autocatalytic oxidation in nitric acid solutions of U(IV) to U(VI). It is converted to gaseous products upon heating, so that it too helps minimize waste solids [7].

In the 1960's, hydrazine-stabilized uranium (IV) was successfully substituted for the chemical reductants previously used. The production of uranium (IV) solutions was accomplished by the electrochemical reduction of the uranyl ion to uranium (V) which disproportionates into U(IV) and U(VI). The disadvantage of the method in which U(IV) is produced in an external reduction facility and incrementally added to the extractor stages is that a three-to ten-fold excess over stoichiometric uranium (IV) must be added to the

process stream [6]. This can result in the isotopic dilution of U-235 in enriched uranium and imposes large volume increases on the process plant [28][6]. Both of these characteristics carry economic disadvantages.

2.3.2. In-situ Electrochemical Reduction

In 1968, an electrolytic partition process, which avoided the need for the introduction of extraneous U(IV), was conceived and subsequently patented by Alfred Schneider, et. al. [29]. The process entailed the in-situ electrolytic reduction of uranium (VI) and plutonium (IV). This technique, employing simultaneous electrochemical reduction and extraction, is referred to as "electrochemical extraction" [15]. The in-situ reactions of importance include the direct reduction of Pu(IV), the direct reduction of U(VI) (both at the cathode), the chemical reduction of Pu(IV) by U(IV) and, if desired, the reoxidation of Pu(III) for subsequent extraction and the destruction of hydrazine (at the anode) [6][3][28]. The applicable reactions are as follows:

$$Pu^{4+} + e \longrightarrow Pu^{3+}$$

$$UO_2^{2+} + 4H^+ + 2e \longrightarrow U^{4+} + 2H_2O$$

$$2Pu^{4+} + U^{4+} \longrightarrow 2Pu^{3+} + U^{6+}$$

$$Pu^{3+} \longrightarrow Pu^{4+} + e$$

$$N_2H_4^+ \longrightarrow N_2 + 4H^+ + 4e$$

Development of the in-situ electrolytic method at the Allied General Nuclear Services (AGNS) reprocessing plant produced the electropulse column, in which mass transfer and electrochemical reduction are carried out simultaneously [24]. An in-situ electrochemical mixer-settler, using a platinum anode (in the anodic chamber) and a titanium casing (to act as the cathode) was demonstrated at the German WAK reprocessing facility [6]. It was developed to technical maturity and successfully operated for about a year [18].

2.3.3. In-line Electrochemical Reduction

The advantages of in-situ reduction can also be realized by providing in-line electroreduction, in which an electrolytic reduction cell is placed between two stages (external to the contactor). This system has the advantage that existing contactors can be modified, rather than having to build an all new facility, as with other in-situ options. Also, the percent reduction of the U and Pu in the stream could be increased over some other in-situ designs (the AGNS electropulse column produced U(VI) to U(IV) reductions of about 3%) [28].

The application of in-line reduction cells would enable the electrochemical reduction technique to be used with centrifugal contactors, since their design precludes the incorporation of electrodes for in-situ reduction. The use of a centrifugal contactor coupled with an electrolytic reduction cell appears to be an attractive way of reprocessing breeder reactor

fuel, since it combines the advantages of the centrifugal contactor (high throughput, high efficiency, critically safe dimensions, etc.) and of electrochemical extraction (avoids salt formation, U-235 dilution, etc.).

2.4. Computer Modeling

2.4.1. Purpose

Modeling of a particular solvent extraction scheme is valuable for determining the number of stages necessary for a given separation, the extraction efficiency for various conditions, etc. In the past, the number of stages required for a particular separation was determined graphically with the McCabe-Thiele diagram [2][3].

With the advent of computers, codes have been developed which take into account the complex combinations of distribution coefficients for several solutes, simultaneous chemical reactions, temperature effects, and transients. These codes are especially useful for modeling the reprocessing of reactor fuel, since they can be used for the continuous on-stream accounting of fissile and fertile isotopes, for determining the maximum stage size to stay within safe criticality limits, to find the optimum parameters (stream concentrations, flow conditions, temperature, etc.), and to determine the effect of transients on criticality considerations [19][2]. They are also valuable for investigating system modifications and for determining the time required to reach steady state.

2.4.2. History

The difficulty of modeling a solvent extraction process increases if simultaneous chemical reactions occur, if the range of concentrations and temperatures is great, and if the number of species to be separated is increased. Modeling of electroreductive uranium-plutonium partitioning, as proposed in this research, requires that the concentrations of U(VI), U(IV), Pu(IV), Pu(III), nitric acid, and hydrazine be calculated at each time interval. The effect on these concentrations by the applicable extraction and redox reactions, both chemical and electrolytic, must also be taken into account [12].

The VISCO computer code was developed at the Karlsruhe Research Center in Germany for the simulation of the transient multi-component extraction in the Purex process. The code was designed in modular form, with care taken to decouple the modules as much as possible. VISCO simulations at the WAK reprocessing plant were found to produce essentially correct results [12].

In the United States, the computer code SEPHIS (Solvent Extraction Process Having Interacting Solutes) was developed at the Oak Ridge National Laboratory. The program models the steady-state and dynamic mass transfer in multi-stage solvent extraction equipment. It has been used extensively for uranium, plutonium, and thorium systems [19]. SEPHIS calculates the step by step changes in uranium, plutonium and nitric acid profiles, enabling evaluation of transient, steady state, start-up and shut-down conditions [3].

SEPHIS is written in FORTRAN IV. The computations are based on a solvent extraction contactor model with a finite series of discrete stages, each containing a "perfect" mixing and settling zone. These zones are described by differential equations, all mass transfer occurs in the mixer, and plug flow is assumed. The code performs the stage calculations using an iterative procedure for each stage in each time interval.

The SEPHIS computer code makes several assumptions and approximations which can be grouped into five main areas [3]:

1. Concentrations in the contactor change relatively slowly.
2. The volumes and flow rates remain constant until changed by the user.
3. The mechanical operation of the contactor conforms with the idealities of the model.
4. Certain chemical effects or conditions are assumed to either exist or not exist.
5. Many heat effects are neglected.

The SEPHIS MOD(IV) version of the program was obtained from ORNL for use in modeling the annular centrifugal contactor in this study. SEPHIS MOD(V), a PC usable version of SEPHIS MOD(IV) compiled by R. Jubin of ORNL, was further modified at Georgia Tech to include uranium (IV) and plutonium (III) distribution coefficients and to model the electrochemical extraction process. This program, designated SEPHIS MOD(V)-GT includes a subroutine to model the reduction cell and the interactions between the cell and the extraction stages.

3. EQUIPMENT AND MATERIALS

3.1. Centrifugal Contactor

A laboratory-size, eight-stage centrifugal contactor, developed at the Argonne National Laboratory, and shown in Figure 3.1, was used throughout this study.

The contactors were fabricated entirely from 304 stainless steel, except for the spindle bearing housing and sleeves which were made of carbon steel [8]. The rotors in the contactor are 20 mm in diameter, hence the designation as a 2-cm size unit. The separation zone lengths were 33 mm and the radial gap between the rotor and housing in the couette mixing zone was 5 mm.

Holes were provided above each flow-tube exit, thus enabling sampling of both phases for each of the eight stages. A syringe was used to withdraw samples during operation. Dry air entering below the spindle and leaving above the aqueous exit port was used to purge the region of acid fumes. Argon gas was used instead of air in some of the experiments with U(IV).

Centrifugal contactors have the advantage of having excellent stage efficiencies, a wide range of operable organic to aqueous ratios and flow rates, rapid start up and shutdown characteristics, favorable geometry and small volumes (important for Pu criticality considerations), and short residence times (important in reducing radiolytic degradation of TBP) [20].

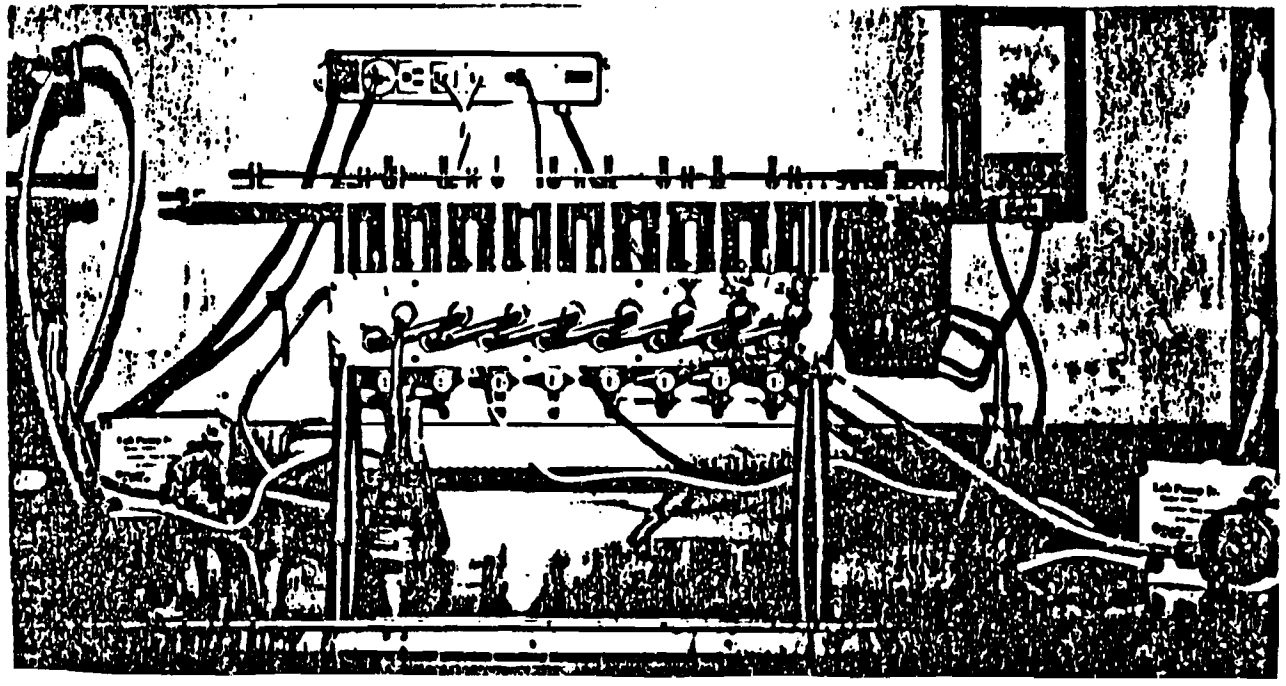


Figure 3.1 Eight-stage Annular Centrifugal Contactor

As shown in Figure 3.2. the two immiscible liquids flow into the annular region between the spinning rotor and the stationary housing, the couette mixing zone. The liquid-liquid dispersion formed in this zone flows by gravity to the rotor inlet in the bottom of the rotor and into the separating zone within the rotor. Here, the dispersion breaks quickly under the influence of the centrifugal force, with the denser aqueous phase moving toward the outside of the separating zone and the less dense organic phase moving towards the center of the separating zone. The separated phases flow over their individual weirs and are ejected by centrifugal force from the rotor into their respective collector rings in the housing. The two liquids leave the collector rings through separate tangential exit ports and proceed to the next stage [5][16].

For multistage operation, units like the one shown in Figure 3.2. are connected together in series. The exiting streams at each stage flow in countercurrent fashion to the next stage. Thus, each rotor acts as a mixer, a centrifugal settler, and a pump.

The multistage contactor capacity at any organic to aqueous ratio is roughly 80% of that for a single stage due to variations in the flow rate between stages. This results in slightly lower mass transfer efficiencies in multistage tests relative to single stage tests. Eight-stage 2-cm annular centrifugal contactors have nominal flow rates of 80 ml/min and mass transfer efficiencies for uranium extraction of at least 85% [5].

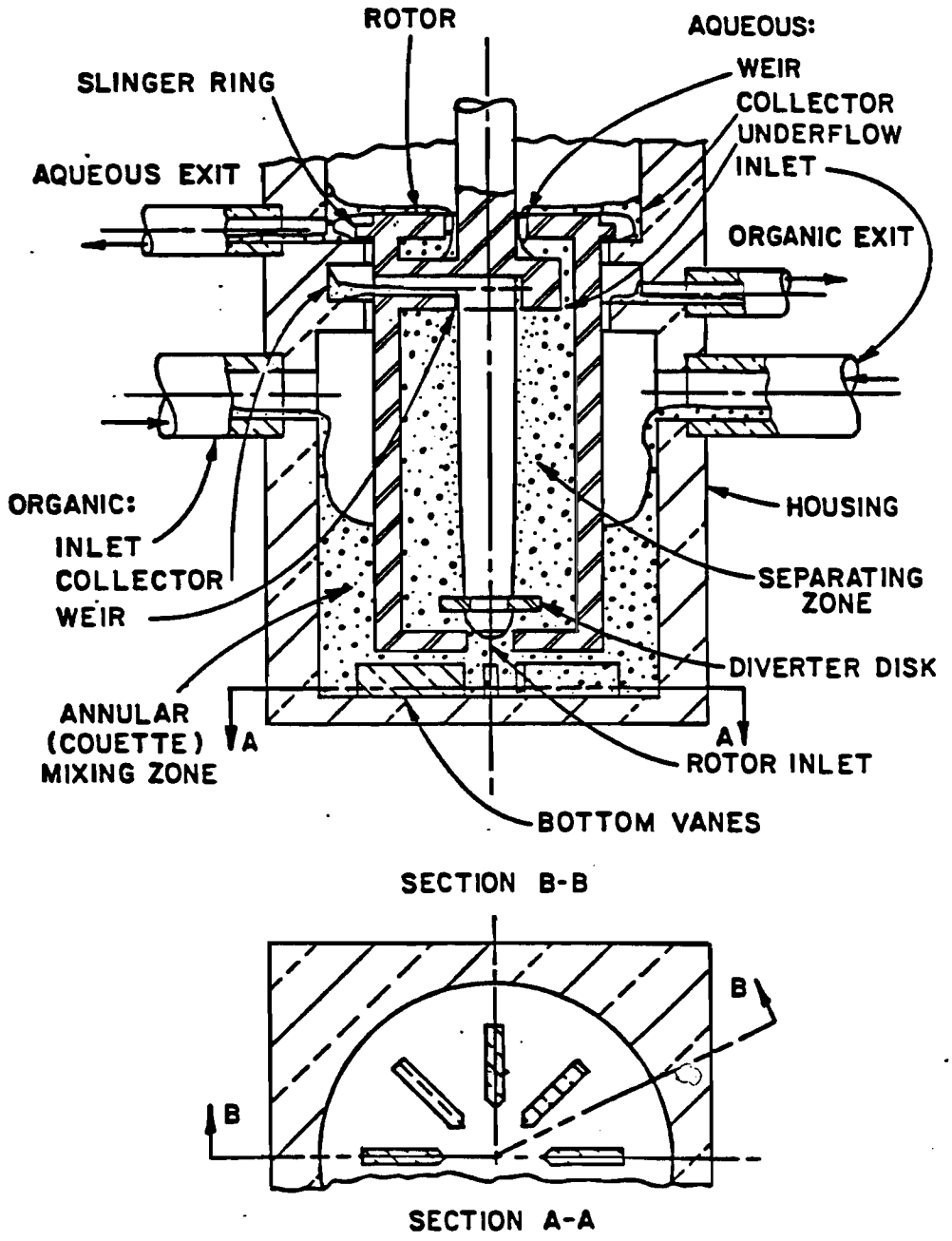


Figure 3.2 Annular Centrifugal Contactor Schematic

Stationary radial vanes under the rotor are provided to decrease the rotational velocity of the dispersion and to allow it to flow by gravity into the rotor. Axial vanes within the rotor cause the liquids inside it to spin at the same rate as the rotor. A diverter disk, inside the rotor and above the inlet, forces the entering dispersion into the middle region of the separating zone. The organic and aqueous weirs were sized so that the dispersion band is located in the separating zone, that is, between the organic weir and the aqueous underflow.

It should be noted that the eight-stage contactor bank had to be scaled down to seven stages for this study, due to unsatisfactory hydraulic performance exhibited by stage eight during hydraulic testing of the contactor.

3.2. Transfer Equipment

The metering pumps obtained from Fluid Metering, Inc. were self-priming, positive displacement types with a fluid temperature limit of 250 degrees Fahrenheit. The flow rates were adjustable from 0.2 ml/min to 30 ml/min with a repeat accuracy of better than 1%.

These pumps performed well, but near the end of this study, the aqueous pump showed some bearing corrosion, presumably due to contact with the 3N nitric acid aqueous solutions.

Initially, only the external feed pumps for the aqueous and the organic streams were used with the contactor. The hydraulic lift of each rotor supplied the gravity head necessary for the countercurrent interstage liquid flow. When

the electrocell was installed between stages 1 and 2, a third pump had to be added directly before the electrocell, since the pressure drop created by the cell interfered with proper flow.

Tygon tubing (1/4 inch) was used for all connections. The tubing used for organic solution transfer showed some embrittlement after about one month. It was reported that contamination of the liquids by leachates from fresh tubing used between the pumps and contactor resulted in an 80% increase in the dispersion number [16]. Because of this, the tubing used in this study was never replaced.

3.3. Electrocell

The "in-line" reduction of uranium (VI) required an electrochemical reduction cell. Originally, an advanced modular reduction cell obtained from Monsanto-Mound Laboratories was to be used. Considerable delays were encountered in obtaining a new membrane for the unit from Hamilton Standard Electrochemical Products, so a small electroreduction cell was designed and built for the study.

Baumgartner and Schmieder reported that no diaphragm was necessary in cells for the reduction of U(VI) in hydrazine-stabilized solutions [6][18][16]. This is due to the fact that after the formation of U(IV) at the cathode it cannot be oxidized at the anode below the potential for oxygen generation. The presence of hydrazine suppresses oxygen generation, since the oxidation of

hydrazine takes place as the preferred anodic reaction [6][18]. Because uranium (IV) reduction and hydrazine oxidation are irreversible processes [18], no separation of the electrode compartments is required for the electrolytic reduction of uranium. The use of diaphragms, which entails undesirable effects such as the generation of heat, an increase in electrical resistance, and possible plugging can be avoided [6][18].

These findings have also been confirmed experimentally by investigators in Beijing, China, and at the Savannah River Plant in the United States [25]. At SRP, a titanium cathode/platinized titanium anode cell with no diaphragm produced a much lower current yield (about 40% reduction). As would be expected for a cell without a diaphragm, the specific hydrazine consumption was much higher than for a cell with a diaphragm.

A small electrocell was designed, built and tested for operation in series with the contactor. The cell anode consists of a pure platinum mesh cylinder, 2 inches high and 1 and 3/4 inches in diameter, surrounding a 1 and 9/16 inches diameter plexiglass core, to minimize the cell's holdup. Fluid enters the cell from the top through a 1/8 inches opening on the center and disperses through eight equally spaced 1/16 inch diameter ports located at the bottom of the cell. (Figures 3.3 and 3.4.) There is a 3/8 inches annular gap around the anode through which the fluid flows upward to the exit port at the top of the cell. At the outer diameter of the annular gap is the cathode, a 2 and 1/2 inches high by 2 and 1/8 inches diameter cylinder of titanium.

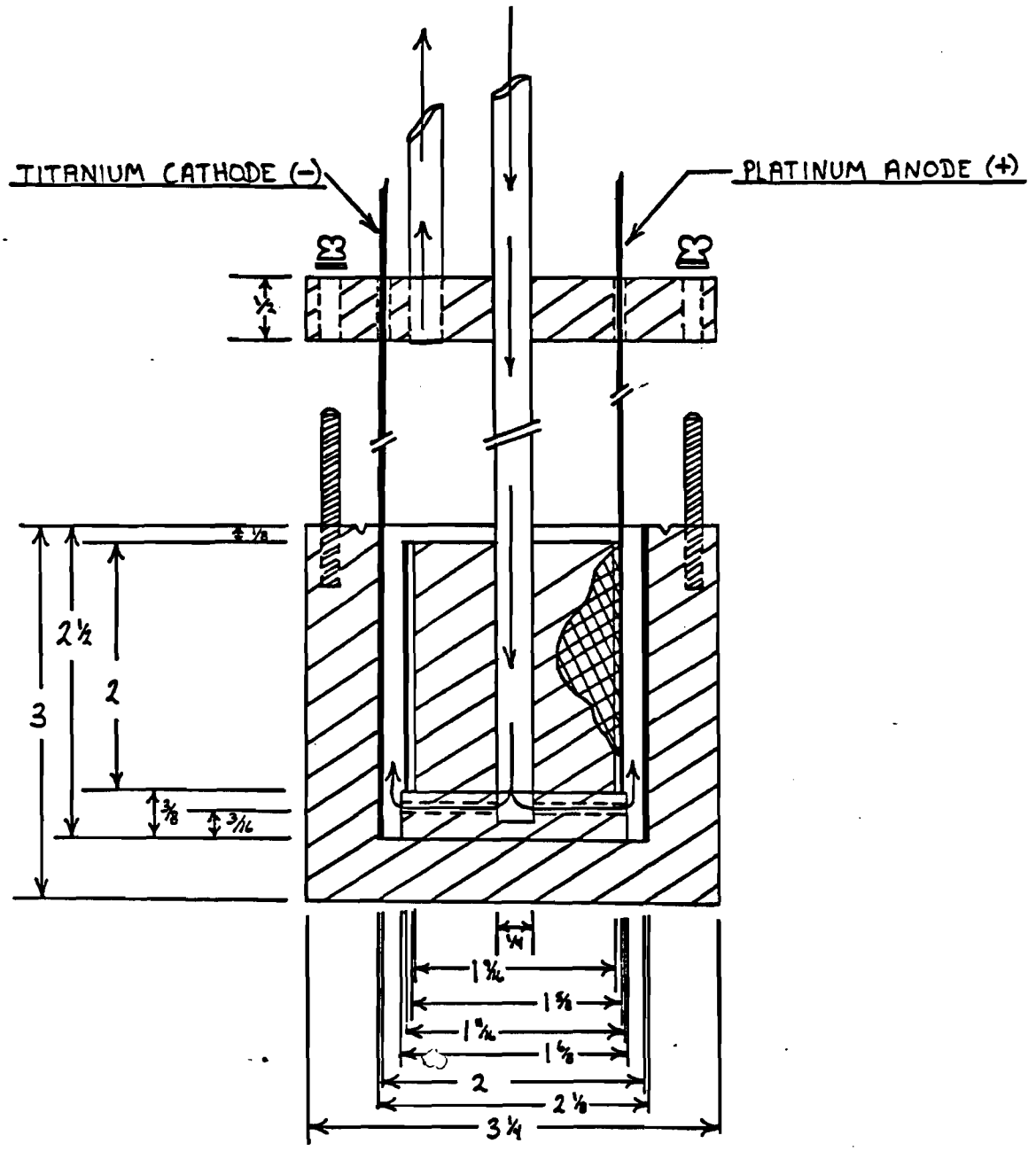


Figure 3.3 Electro-reduction Cell - Side View

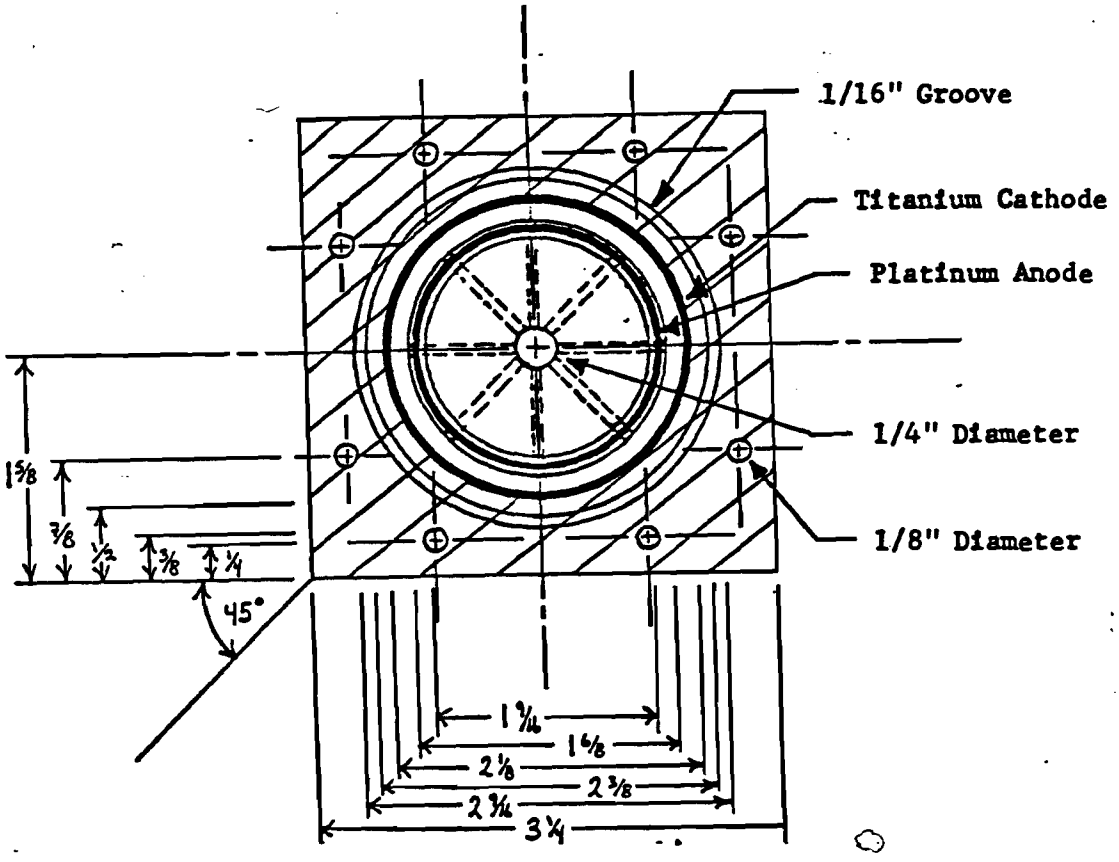


Figure 3.4 Electro-reduction Cell - Top View

The cell is contained in a 3 and 1/4 inches square block of Plexiglass, 3 and 1/2 inches tall. A rubber O-ring is placed between the cell and its lid so that the fluid entering the cell is forced out of the cell lid through a single 1/4 inch exit tube. The cell has a cathode area of 92.7 cm^2 and a holdup volume of 47.6 cm^3.

Tests showed that U(IV) can be produced in a continuous mode at flow rates necessary for operation in tandem with the centrifugal contactor. A maximum reduction of U(VI) to U(IV) of about 30% was achieved for an original uranyl nitrate solution of 0.157 M uranium (VI).

The constant current voltage supply used was an Electro-Analysis Apparatus made by the Eberbach Corporation, with a voltage range from 0 to 10 volts and an adjustable current from 0 to 10 amps.

3.4. Evaporator

An electronically controlled evaporator was designed for this project. The evaporator consisted of a 1000 ml, triple-necked distillation flask, a heating mantle and variable voltage supply, a water-cooled glass condenser and a 1000 ml distillate flask. The apparatus was designed with temperature and pressure controls to prevent overheating. This was necessary, since explosions of "red oil" have occurred in heated solutions containing TBP, nitric acid and uranyl nitrate, [3][36].

Two THERMO-O-WATCH controllers, made by the Instruments for Research and Industry Company, were used. Each THERM-O-WATCH controller consisted of a sensing head which clipped onto the thermometer or manometer in order to sense the rise or fall of mercury and an amplifier, which controlled two electrical outlets. Once the sensing head was properly adjusted, a slight change in the mercury column height altered the electrical capacitance in the sensing head causing the amplifier outlets to turn on or off [33].

Additional features included a High Efficiency Particulate-Free Air (HEPA) Capsule located between the evaporator and the solenoid valve (to avoid any possibility of contaminating the solenoid or vacuum pump), a hose clamp between the solenoid and the vacuum pump (to adjust the suction of the vacuum pump) and two blast shields on either side of the evaporator. A schematic drawing of the system is in Figure 3.5.

3.5. Spectrophotometer

A Bausch and Lomb Spectronic 21 spectrophotometer was used for the colorimetric analyses. This spectrophotometer had a wavelength range from 310 to 1000 nanometers (nm).

All aqueous samples were contained in square, disposable cuvets (Ultra-VU microcuvets), made of poly-styrene. All organic samples were placed in round Bausch & Lomb glass cuvets due to the fogging effect the organic samples had on plastic cuvets.

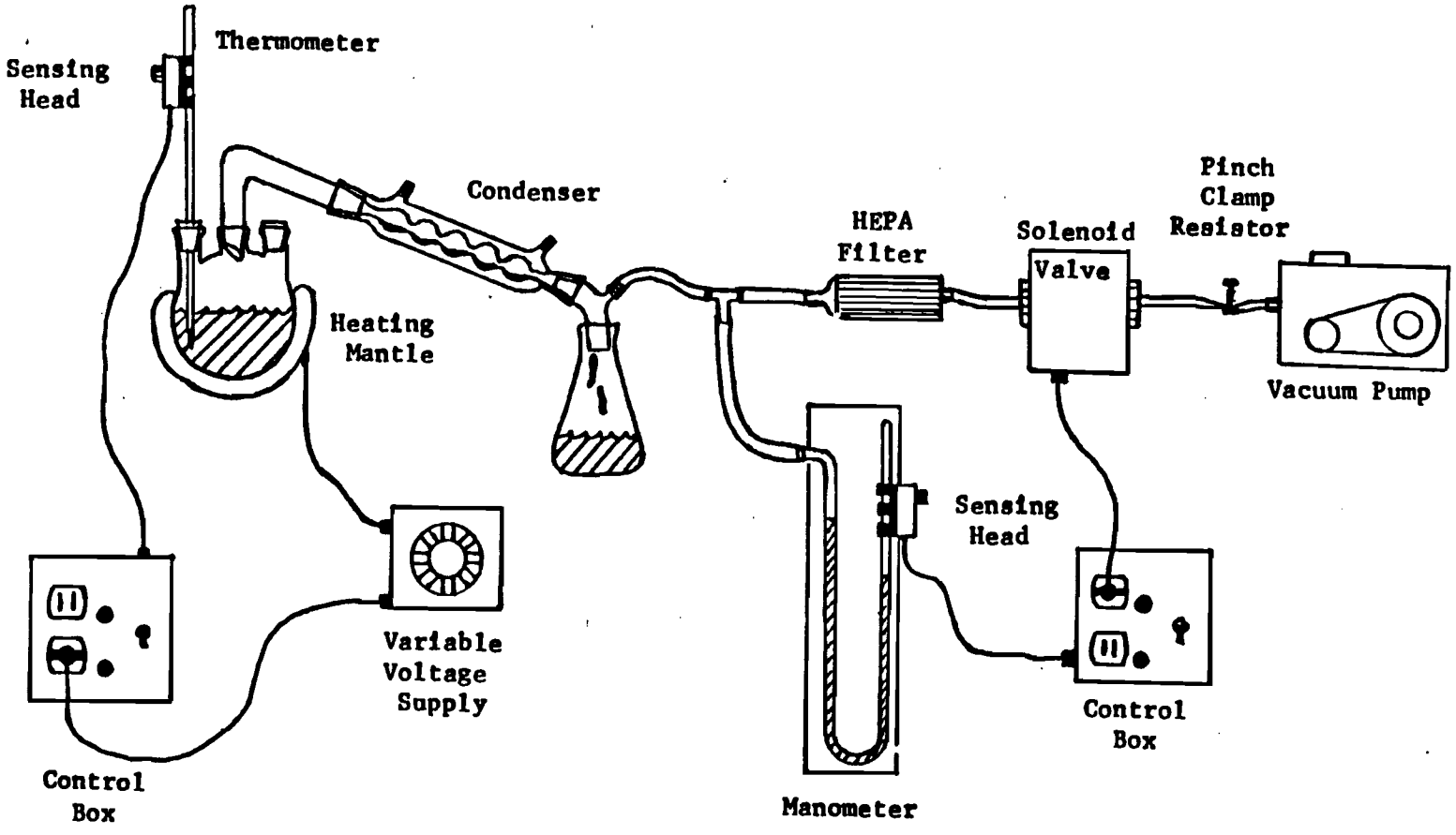


Figure 3.5 Evaporator Apparatus

3.6. Chemicals

3.6.1. Uranyl and Uranous Nitrate

The uranyl nitrate solution had been previously used [8] and was purified by batch extractions/strippings. All experimental tests using the centrifugal contactor or the electrocell were run with this solution.

Uranyl nitrate standards, used for the calibration of analytical techniques, were produced by dissolving carefully weighed amounts of reagent grade uranyl nitrate hexahydrate ($UO_2(NO_3)_2 \cdot 6H_2O$) crystals in nitric acid.

All uranous nitrate solutions, whether for use as analytical standards or for experimental runs, were produced by reducing uranyl nitrate with the electrocell. Uranium (IV) reoxidizes easily to uranium (VI) in nitric acid solutions and increases in acidity cause increases in the rate of U(IV) oxidation [6][14][25]. To prevent reoxidation, hydrazine stabilizer was added in all uranium (IV) - containing solutions.

3.6.2. Hydrazine

Solutions of uranous nitrate, stabilized with hydrazine, were found to be a successful substitute for chemical reductants in the Purex process in the 1960's [26]. Hydrazine is a stabilizer for U(IV) since it minimizes the presence of nitrate ions.

Hydrazine acts as a salting agent in uranyl nitrate, increasing the amount of U(VI) and U(IV) extracted into the organic phase [11]. The presence of

hydrazine has also been found to promote the rate of reduction of U(VI) [27][25]. Hydrazine can be completely converted to gaseous nitrogen oxides and water.

All uranous nitrate solutions were stabilized using hydrazine concentrations of between 0.2 M and 0.5 M. Hydrazine does not extract appreciably, into the organic phase; however, hydrazoic acid (N_3H) produced from hydrazine does extract. Under certain circumstances, N_3H can decompose violently.

3.6.3. Tributyl Phosphate

Tributyl phosphate (TBP) is the most widely used extractant for uranium and plutonium [8]. The formation of uranium complexes with TBP occurs primarily by the following reaction [23]:

$$UO_2^{++}(aq) + 2NO_3^-(aq) + 2TBP(org) \rightleftharpoons UO_2(NO_3)_2 \cdot 2TBP(org)$$

In this research a solution of 30% TBP and 70% Norpar-12 (a hydrocarbon diluent obtained from Exxon Chemical Corporation) was used as the organic solvent.

Tributyl phosphate is an ester of phosphoric acid which gradually undergoes degradation when exposed to high radiation in the presence of water and nitric acid. The reactions involved are as follows [3]:

$$PO(C_4H_9O)_3 + H_2O \longrightarrow H(C_4H_9O) + PO(OH)(C_4H_9O)_2$$

$$PO(OH)(C_4H_9O)_2 + H_2O \longrightarrow H(C_4H_9O) + PO(OH)_2(C_4H_9O)$$

$$PO(OH)_2(C_4H_9O) + H_2O \longrightarrow H(C_4H_9O) + PO(OH)_3$$

Radiolytic damage is especially important in the Purex process for the reprocessing of reactor fuel which involves intense radiation. This creates the need for minimizing the residence time of the organic liquids in the contactor. The effect of radiolytic degradation is an increase in uranium and plutonium losses, because the salts formed with dibutyl phosphate (DBP) and monobutyl phosphate (MBP) do not decompose during stripping. Moreover, MBP and DBP extract under TBP stripping conditions (i.e. water or low acid concentrations), and strip under TBP extracting (i.e. high acid concentrations) conditions.

3.6.4. Analytical Reagents

All chemicals used were certified reagent grade obtained from the Fisher Scientific Company. These chemicals included: 85% phosphoric acid, concentrated nitric acid, 3% hydrogen peroxide, sodium hydroxide solution (N/10), potassium biphthalate, potassium dichromate, ferric chloride, ferric ammonium sulfate, ferrous ammonium sulfate and the indicators congo red, methyl orange, ferroin, methyl red solution (0.02%), phenolphthalein, and sodium diphenylamine sulfonate.

4. EXPERIMENTAL PROCEDURES

4.1. Safety Precautions

The necessary safety measures were taken when working with radioactive materials. This included wearing a thermoluminescent dosimeter (TLD), monitoring the work areas with a pancake type Geiger counter, and the isolation of all radioactive wastes.

4.2. Centrifugal Contactor Operation

Difficulties were encountered initially with the hydraulic performance and the sampling of the contactor's individual stages. These problems were resolved and the contactor's stage-wise performance proved satisfactory.

4.2.1. Start up

Before the starting of an extraction or stripping experiment, the contactor was drained of any fluids remaining from previous experiments through the drainage plugs at the bottom of each stage to remove any precipitates or corrosion products.

First, the aqueous pump was turned on. After approximately 10 to 20 minutes, depending on the aqueous flow rate, the aqueous solution reached the exit stage. The organic pump was then turned on. If the aqueous is not sent all the way through the contactor before the organic pump is started there is an increase, by a factor of 2 to 3, in the time it takes to reach steady state [8]. This is due to the resistance to phase inversion, from organic-continuous to aqueous-continuous, in the couette mixing zone. In contrast, the aqueous-continuous to organic-continuous phase inversion proceeds promptly [5]. The contactor bank was run at a speed of 6200 rpm throughout this study.

4.2.2. Sampling Techniques

After clean phases (less than 1% phase contamination) were observed leaving the organic and aqueous exit stages, samples were taken at each exit point at approximately 5 minute intervals and analyzed to determine when

steady state was reached. This was done for all runs and equilibrium was normally reached after about 20 minutes. Therefore, no stage samples were taken subsequently before at least 30 minutes had elapsed.

Samples were obtained directly at the mouth of the aqueous and organic stage exit tubes through sampling holes in the top of the contactor housing using a 10 ml syringe. Two of the contactor's sampling holes were plugged and could not be reopened. To allow on-line sampling of these two exit stages the stainless steel flow tubes, on the outside of the contactor, were replaced with tygon tubing fitted with T-connectors for access to the stage exit flow.

With stage sampling occurring on-line it was desirable to minimize the disturbances to subsequent stages due to the withdrawal of fluid (usually about 5 ml) from a stage. The procedure adopted was to withdraw samples starting at the exit stages and, alternating between aqueous and organic streams, work towards the entrance stages.

4.3. Recycling of Uranium

The initial purification of the uranyl nitrate solution and all subsequent cleanup of the solution was accomplished by batch operations. The highly dilute uranyl nitrate solutions, resulting from the batch stripping stage, were then concentrated using the evaporator.

4.3.1. Batch Purification

The procedure for purifying uranyl nitrate solutions was to mix them vigorously in a separatory funnel with a 30% TBP, 70% Norpar organic solvent in an O/A ratio of about 4 to 1. The aqueous raffinate and the organic extract were then drained into separate bottles and the process was repeated until all the original uranyl nitrate had been contacted. The resulting solutions were the organic, containing uranium and a small quantity of free nitric acid, and the depleted aqueous, containing small amounts of the unextracted uranium and other impurities. The aqueous waste stream from this step was concentrated in the evaporator and stored as concentrated waste.

The organic extract could then be used in stripping or simulated partitioning runs. If uranium in the aqueous phase was required, the organic was batch stripped. By contacting the organic with distilled water at an O/A ratio of about 1 to 4 a pure, but very dilute, uranyl nitrate solution was produced. Thus, in purifying one liter of concentrated uranyl nitrate, 16 liters of dilute uranyl nitrate was produced. This solution was concentrated with the evaporator.

4.3.2. Evaporator Operation

Concentrated uranyl nitrate, nitric acid and TBP mixtures are known to form hazardous "red oil" complexes, which can lead to violent explosions, when heated to temperatures exceeding 130°C [3]. To decrease the presence of TBP - hydrocarbon compounds necessary for "red oil" formation the aqueous

solution was always drawn from the bottom of its container, since it is heavier, in the presence of organic compounds. To prevent the temperature of the solution from reaching 130°C the system was operated at reduced pressure. The temperature and pressure were maintained at around 70°C and 170 mm Hg, respectively, to stay well within the safe region of operation. In addition, the operator was present at all times the evaporator was turned on to observe that there was no visible "red oil" formation in the apparatus and that the solution was never boiled to dryness. As a final precaution, blast shields were placed around the evaporator to minimize any injuries due to a possible explosion.

Two independent methods were used to control the evaporator's temperature. The variable voltage supply was controlled by a sensing head attached to the distillation flask's thermometer and set at 110°C.

The temperature was also maintained by controlling the pressure of the closed system. The pressure of the evaporator was monitored by a manometer. A vacuum pump was used to maintain the reduced pressure, thus lowering the boiling temperature of the solution. The reduced pressure of the evaporator was controlled by an electronic solenoid valve placed between the system and the vacuum pump. The solenoid was plugged into a THERMO-O-WATCH amplifier and its sensing head was attached to the manometer. An increase in pressure would open the solenoid valve and allow the vacuum pump to reduce the system's pressure.

Care had to be taken that the opening of the solenoid did not reduce the system pressure too suddenly or boilover of the solution into the condenser would occur, contaminating the distillate. To ensure that the pressure reduction resulting from the opening of the solenoid valve was a gradual one, a hose clamp was placed on the vacuum hose leading to the evaporator and adjusted until the problem with sudden boilover was alleviated.

The distillate was removed and stored in a large holdup container which was tested for activity before disposal. The distillate never showed any activity above background.

4.4. Analytical Techniques

Since a large number of samples had to be analyzed for each experiment it was desirable to use analytical methods which were simple and not time-consuming. Colorimetric analyses were employed whenever possible. Analytical methods were needed for U(VI) and U(IV) concentrations in aqueous and organic solutions and for the free acid content.

4.4.1. Determination of Free Acid in Aqueous Solutions

The determination of free acid in uranyl nitrate solutions was done by the peroxide precipitation method [4]. This involved the addition of hydrogen peroxide to a solution or uranyl nitrate, causing the precipitation of uranium in accordance with

$$UO_2(NO_3)_2 + H_2O_2 \longrightarrow UO_4 + 2HNO_3$$

The total acid, formed from this reaction and the free acid originally present, was then titrated with a standard 0.1N sodium hydroxide solution to a pH of about 4.5. The free acid was found by subtracting the acid generated during the peroxide precipitation (calculated from the uranium concentrations) from the total acid determined by titration.

$$\text{Free Acid (N)} = \frac{[\text{NaOH(N)}] \times [\text{NaOH (ml)}]}{[\text{sample vol (ml)}]} - 2\,[UO_2(NO_3)_2]$$

Three indicators which change color in the 4.5 pH range were investigated. Congo red, which changes from blue (pH 3.0) to red (pH 5.0), was found to give very poor results due to an unclear color change. Methyl orange, red (pH 3.2) to yellow (pH 4.4), gave better results but the end point was not sufficiently sharp and this resulted in an average error of only -.4% but a high standard deviation of 3.4%.

Methyl red, red (pH 4.2) to yellow (pH 6.2) was found to give a much sharper color change. The average error was +.18% with a standard deviation of 1.33%. The results for methyl red, as well as the other two indicators, are presented in Appendix A, Table A.1. Methyl red indicator was therefore used in all free acid measurements of uranyl nitrate. Standardization of the 0.1 NaOH was done with potassium biphthalate using a phenolphthalein indicator [30].

4.4.2. Determination of Free Acid in Organic Solution

The nitric acid in the organic phase was stripped into pure water and the acidity of the aqueous phase was determined by the peroxide precipitation method described in section 4.4.1. A correction was included for an assumption that 5% of the free acid remained in the organic phase.

4.4.3. U(VI) in Aqueous and Organic Solutions

The spectrophotometric determination of U(VI) in aqueous and organic solutions was found to be adequate. This method required little sample preparation, was fairly accurate (±5%) and was not too time consuming. Since the spectrophotometric reading for a given sample was a relative number, calibration curves had to be generated to correlate the instrument readings with actual standards. Each correlation curve produced was checked against independent test samples to obtain the average error and standard deviation for each curve.

U(VI) Absorption Spectra

The maximum absorption band usually listed for the determination of U(VI) is at a wavelength of 410 nm [9]. This varies for different instruments, so absorption spectra were plotted for two different U(VI) concentrations in both the aqueous and organic phases.

Aqueous solutions of uranyl nitrate (0.103 M and .025 M) in 3 N nitric acid were tested at wavelengths from 350 to 520 nm in 10 nm increments (finer

increments were used in the region of highest absorption). The scale was recalibrated for each wavelength setting using a 3 N nitric acid blank. In both cases the spectrophotometer was most sensitive to U(VI) at a wavelength of 415 nm; therefore all U(VI) determinations were done at this wavelength. The results are shown in Figure 4.1. Disposable plastic cuvets were used for all aqueous samples and blanks.

The absorption spectra for organic samples of 0.117 and 0.029 M uranyl concentrations were obtained by the same method and over the same wavelength range as the aqueous spectra. Bausch & Lomb round glass cuvets were used for all organic samples and blanks. Again, the highest sensitivity to U(VI), for both solutions was found at a wavelength of 415 nm. This is verified by Yu Novikov who lists the maximum U(VI) absorption at a wavelength of 416 nm in a 30% TBP medium [9]. The absorption spectra in the U(VI) organic solutions are presented in Figure 4.2.

U(VI) Spectrophotometric Correlations in Aqueous

The spectrophotometric absorption of U(VI) in aqueous solutions was correlated with known (U(VI) standards over the entire concentration range measurable with the instrument. Nitric acid concentration affects the absorption of U(VI), therefore standards were prepared at O N (distilled water), 1 N and 3 N nitric acid concentrations [9]. Standards were prepared by carefully weighing out uranyl nitrate hexahydrate crystals to produce concen-

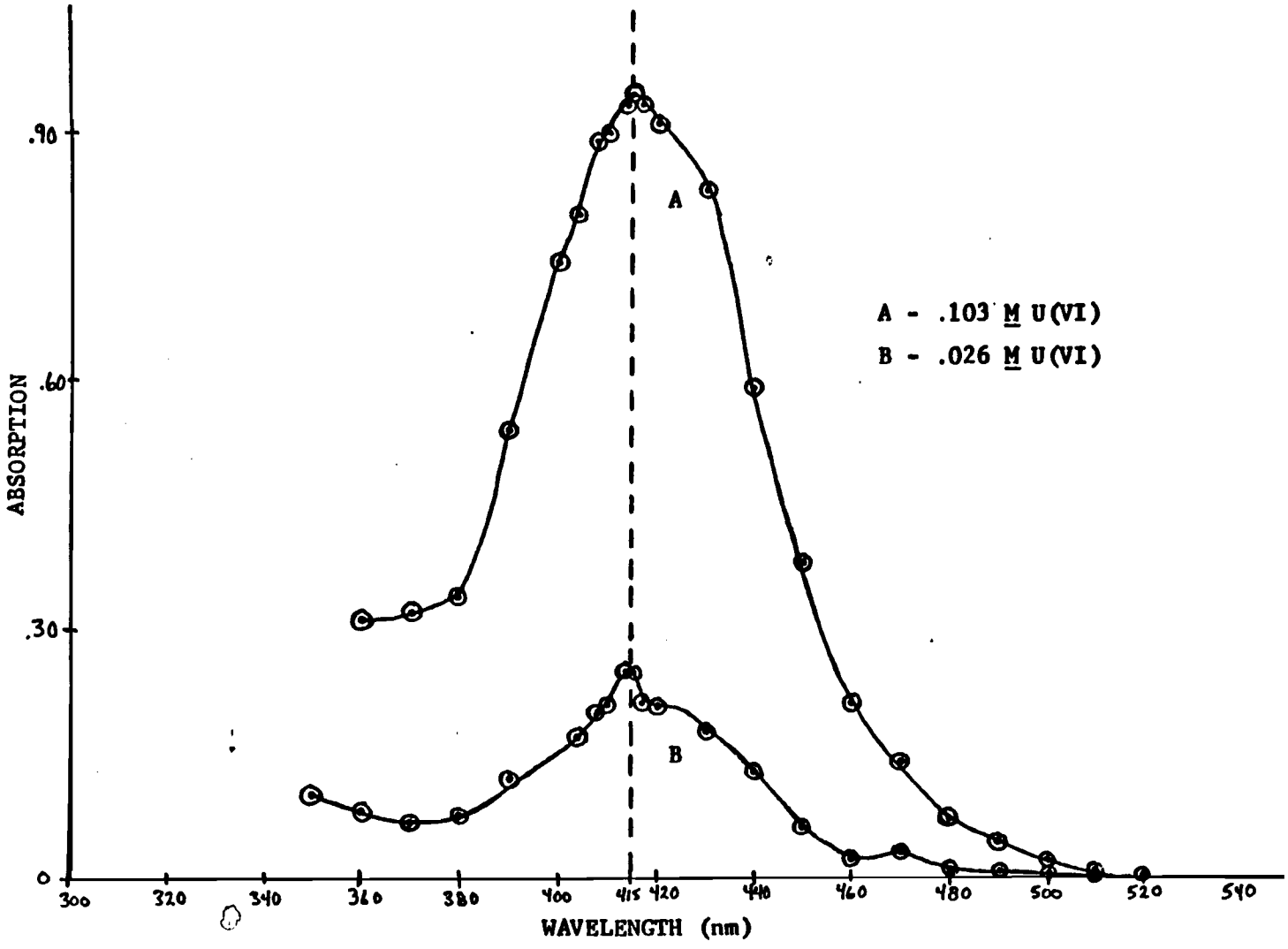


Figure 4.1 U(VI) Absorption Spectra in Aqueous

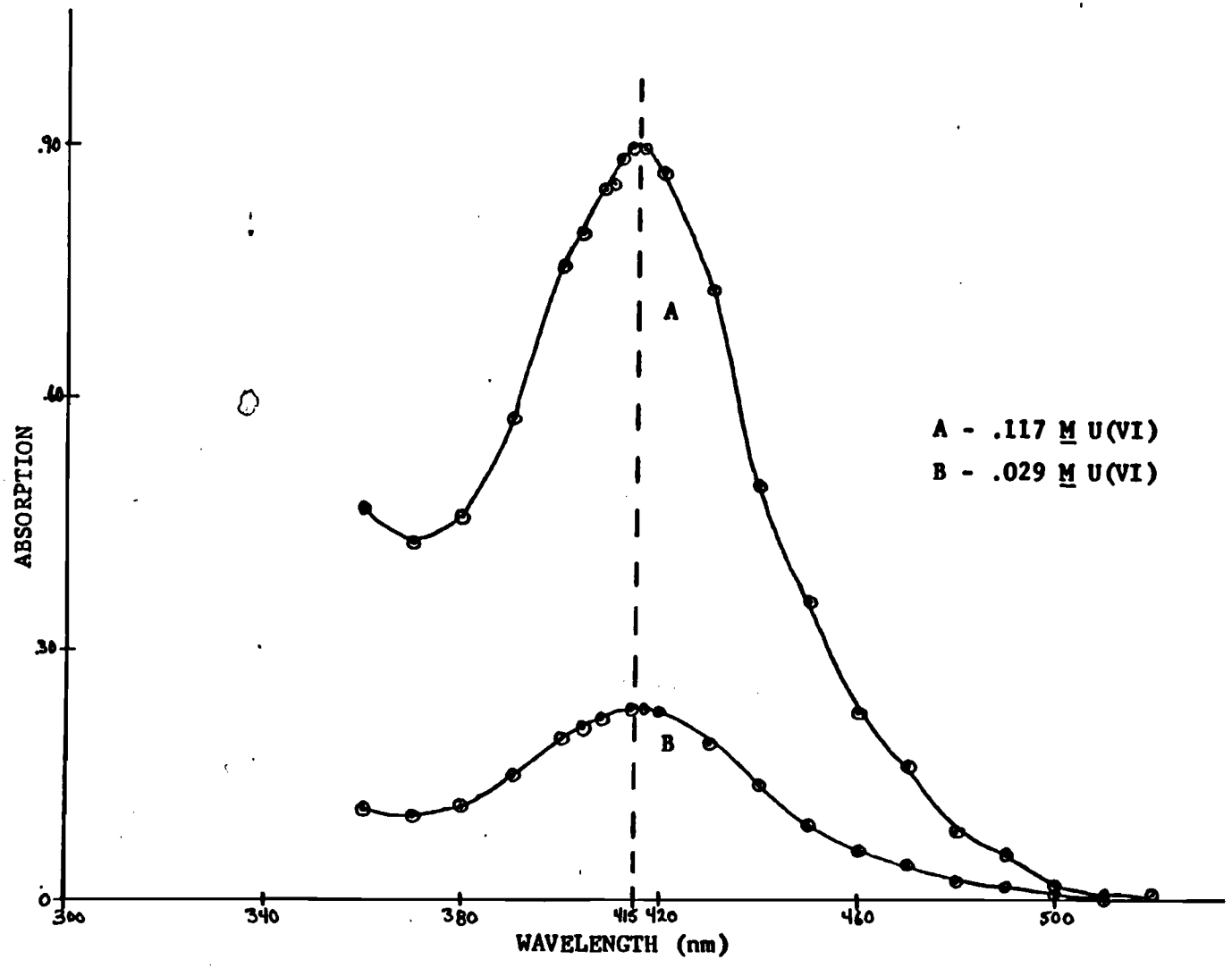


Figure 4.2 U(VI) Absorption Spectra in Organic

trations in the range of about .01 M to .1 M U(VI). These crystals were then diluted in either O N, 1 N, or 3 N nitric acid. The absorption of each standard was checked three times (at 415 nm) and the readings averaged. The spectrophotometer was recalibrated for each new sample using O N, 1 N or 3 N nitric acid blanks.

A least squares fit program was used to generate the correlation equation and the corresponding correlation coefficient for each acidity. In all cases the relationship between the uranium concentration of the sample and its corresponding absorption was found to be linear. The tabulated results, giving the data points used, absorption readings, correlation equations and corresponding correlation coefficients, are presented in Table A.2. The correlation curves for the three acidities are plotted in Figure 4.3. It is clear from these curves that the acidity of the uranyl nitrate sample does have an effect on absorption, particularly as the U(VI) concentration increases.

As a check on these spectrophotometric correlations an independent batch of samples was prepared for each acidity. The correlation curve error data are presented in Table A.3. The accuracy for all three equations was proven to be very satisfactory, with all sample errors within 3.5% and standard error deviations of about 2% for each of the curves.

U(VI) Spectrophotometric Correlations in Organic

The methods used for correlating U(VI) absorption measurements in organic solutions were identical to the procedures used for aqueous samples.

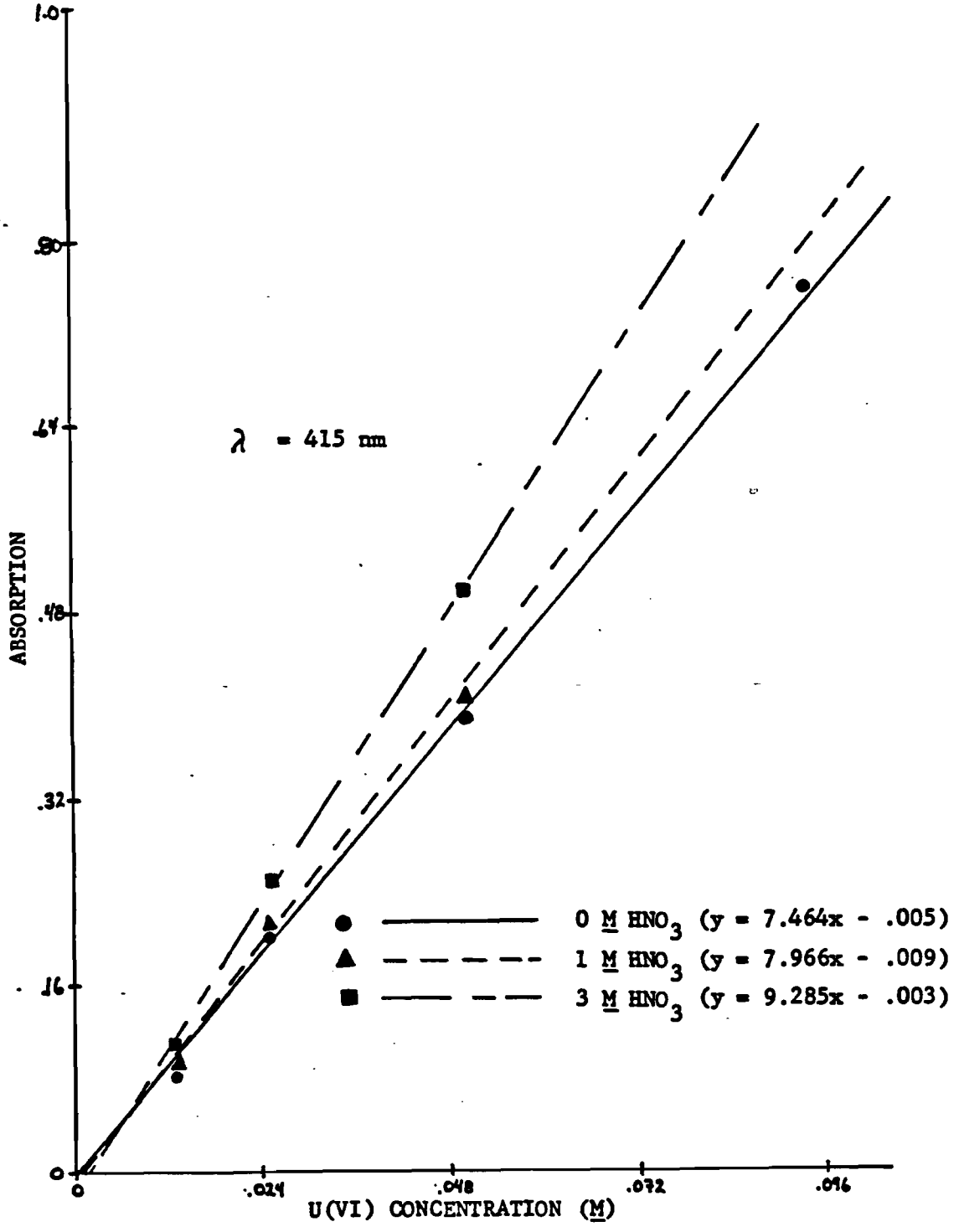


Figure 4.3 U(VI) Colorimetric Correlations - Aqueous

Organic standards used were obtained by small batch equilibrations of 30% TBP in 70% Norpar solutions with uranyl nitrate solutions. The amount of uranium in the organic samples was determined by taking the difference between the aqueous uranium content before and after the extraction. The organic standards were calibrated against clean 30% TBP solutions.

The data points used, correlation equation, and correlation coefficient for the curve are given in Table A.2. The calibration curve for U(VI) in organic solutions is presented in Figure 4.4.

Replicate determinations gave an average error of 7.93% and a standard deviation for the correlation of 3.09%.

Higher errors, compared to the aqueous correlation, were expected since the determination of organic U(VI) content in the samples was done by difference.

4.4.4. Determination of U(IV) in Aqueous Solutions

Potassium Dichromate Method

The potassium dichromate method with a diphenylamine indicator, was first tested for U(IV) determination [9]. An advantage of this method is that potassium dichromate in water is stable indefinitely [10], making it the primary standard [32].

U(IV) is first oxidized to U(VI) by adding a 4% solution of ferric chloride in the presence of phosphoric acid [9][10]. The equivalent amount of Fe(II) formed in the solution is then titrated with 0.1 N potassium dichromate solution in the presence of phosphoric acid.

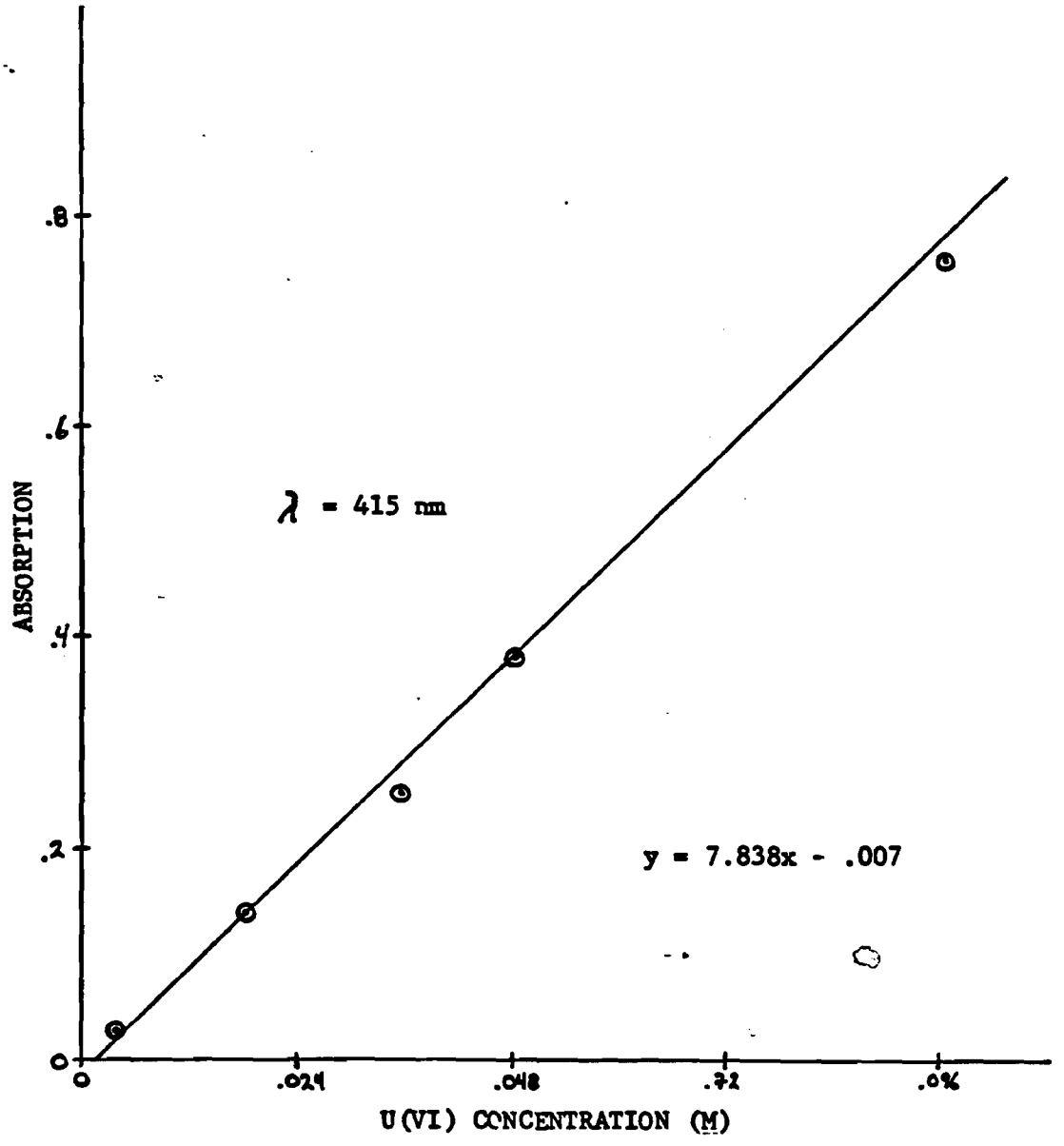


Figure 4.4 U(VI) Colorimetric Correlation - Organic

Several "blank" (no uranium) samples were tested to determine the effects of nitric acid or hydrazine. The initial blank tested was 2.8 N nitric acid only. Next, several samples of 2.8 N nitric acid and 0.2 M hydrazine were tested.

The nitric acid solution alone gave a very clear gray-green to purple color change; however, the samples containing hydrazine gave no clear indication of the equivalence point and the solution changed to dark brown. The presence of reducing agents, such as hydrazine, interferes with this analysis [32]. This may have been because the analysis was based on oxidizing U(IV) to U(VI), which the hydrazine is meant to prevent.

Ceric Sulfate Method

The ceric sulfate titration method [9] was then tested. In a H_3PO_4 and H_2SO_4 medium, Ce(IV) (as ceric ammonium sulfate) oxidizes U(IV) as follows [31]:

$$U(IV) + 2Ce(IV) = U(VI) + 2Ce(III)$$

The end point was detected by the indicator ferroin (red to colorless), which was oxidized by the Ce(IV), only after there was no more U(IV) left to be oxidized.

The ceric ammonium sulfate was first standardized by ferric ammonium sulfate (a secondary standard) since Ce(IV) tends to be unstable. The ferric ammonium sulfate was standardized in turn by potassium dichromate, since FAS is unstable but potassium dichromate is stable.

Interference due to the presence of hydrazine was a concern since it can, under certain conditions, consume Ce(IV). Under the conditions chosen for this analysis, however, this reaction does not occur.

A correction was required for the slight amount of Ce(IV) consumed by the ferroin indicator, and to account for the presence of nitric acid and hydrazine in the test samples. Three 10 ml "blanks", containing nitric acid and hydrazine at concentrations of 2.8 M and 0.2 M respectively, were titrated with 0.0058 N ceric sulfate and ferroin indicator. No uranium was present.

It took an average of 0.25 ml of the 0.0058 N ceric sulfate to account for the nitric acid, hydrazine and ferroin present in the samples. Thereafter the volume of ceric sulfate required for the actual 10 ml aqueous U(IV) samples was reduced by 0.25 ml to account for these factors. All U(IV) samples used for the spectrophotometric correlation data were analyzed the same day the ceric sulfate concentration was determined to avoid any possible ceric sulfate stability problems.

The method proved satisfactory and no problems due to the presence of hydrazine or nitric acid were observed. The ceric sulfate method was used for U(IV) in organic as well as aqueous solutions.

4.4.5. U(IV) in Aqueous and Organic Solutions

U(IV) Absorption Spectra

The maximum absorption band usually listed for the determination of U(IV) is at a wavelength of 640 nm [9]. An absorption spectrum was plotted for a sample with 0.18 M U(IV), 2.8 N nitric acid and 0.2 M hydrazine.

The sample was tested at wavelengths from 635 nm to 655 nm in 5 nm increments (finer increments were used in the region of highest absorption). The spectrophotometer was recalibrated for each new wavelength with a 2.8 N nitric acid, 0.2 M hydrazine blank. The spectrophotometer was found to be most sensitive to U(IV) at a wavelength of 648 nm; therefore, all U(IV) determinations were done at this wavelength. The U(IV) absorption spectrum in an organic medium is shown in Figure 4.5.

U(IV) Spectrophotometric Correlations in Aqueous

Ions of uranium (IV) absorb light in the same region of the spectrum (415 nm) as those of uranium (VI). Uranium (IV), however, can be determined at longer wavelengths (648 nm) [9]. Thus, the concentration of U(IV) in samples containing both U(IV) and U(VI) was determined solely by testing the sample at a wavelength of 648 nm. The concentration of the uranium (VI) ion, however, was determined using differential spectrophotometry once the U(IV) concentration in the sample was found. The accuracy of uranium (IV) determination depended on the U(IV)/(U(VI) concentration ratio. The smaller this ratio, the higher the accuracy of the U(VI) determination [9].

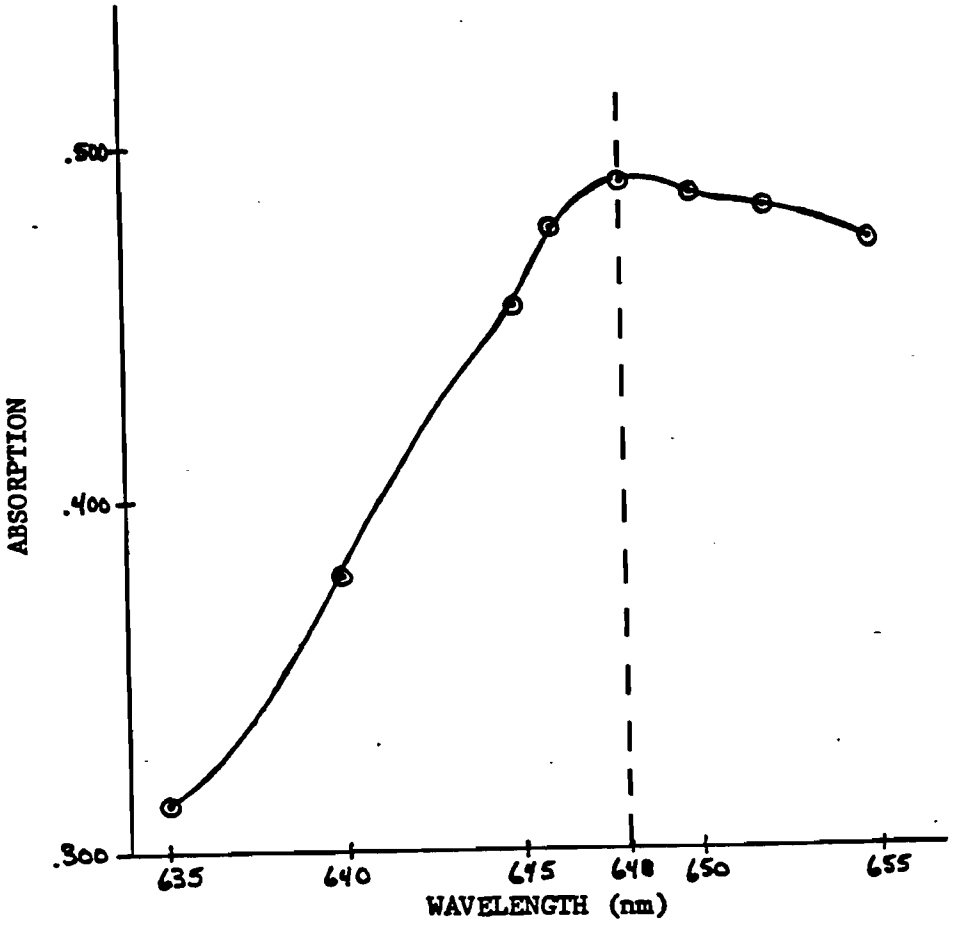


Figure 4.5 U(IV) Absorption Spectra in Organic

The correlation of U(IV) concentration versus absorption readings, at 648 nm, was accomplished in a similar manner to that used for the U(VI) correlation curves. Five samples of varying U(VI) concentrations, each with a content of 2.8 N nitric acid and a 0.2 M hydrazine, were run through the electroreduction cell.

Each sample was analyzed for uranium (IV) content using the ceric sulfate method described in Section 4.4.4. Samples of 10 ml were used in all titrations. Spectrophotometric absorption readings were taken for each of the solutions. An average of three values was used for determining the U(IV) concentration and absorption readings. The samples varied in U(IV) content between 0.0013 M and 0.0057 M and in absorption readings between 0.024 and 0.148, respectively.

The correlation between the U(IV) content of the samples and their corresponding spectrophotometric absorptions was found using a least squares fit program. The experimental data, the corresponding correlation equation, and correlation coefficient are given in Table A.4. A plot of the curve is presented in Figure 4.6.

A separate, independent batch of test samples was prepared and analyzed for uranium (IV) content. These were checked against the U(IV) concentration found using the spectrophotometric correlation for U(IV). The mean error was -0.8% with a standard deviation of 3.95%. The data are summarized in Table A.5.

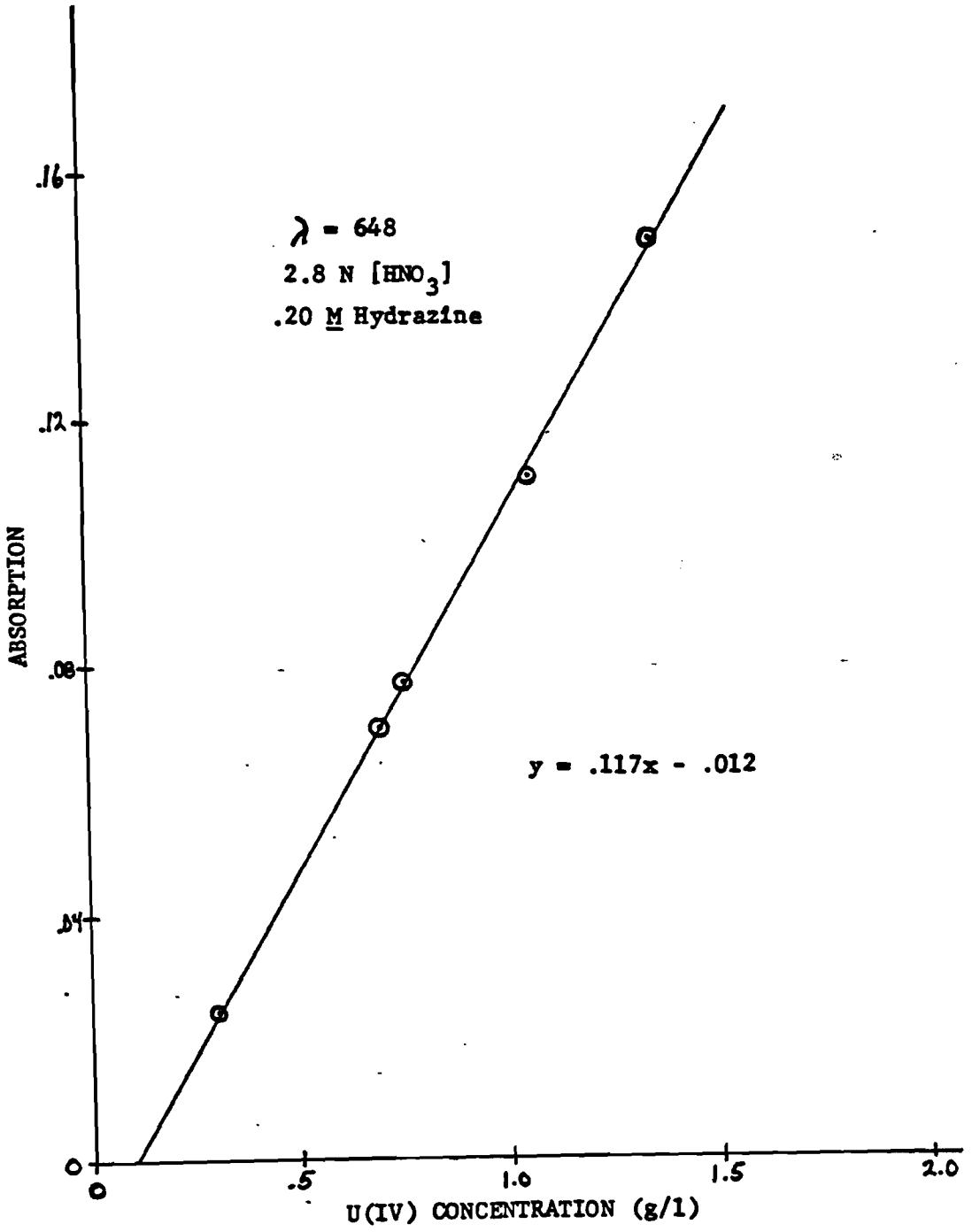


Figure 4.6 U(IV) Colorimetric Correlation - Aqueous

U(IV) Spectrophotometric Correlations in Organic

Small batch extractions were utilized to transfer the uranium (IV) in aqueous samples into clean solutions of 30% TBP and 70% Norpar. The absorption of the samples was read at 648 nm in glass, Bausch & Lomb cuvets and calibrated against a clean 30% TBP sample. The uranium (IV) content was found, for 10 ml samples, using the same ceric sulfate method used for U(IV) in aqueous determinations.

The data is presented in Appendix B, Table B.7, along with the correlation equation and its correlation coefficient. A least squares fit was used to plot the data in Figure 4.7.

For replicate samples, a mean error of -2.6% and a standard deviation of 5.7% were obtained. The data are listed in Table A.5.

Colorimetric methods for the determination of uranium (IV) and uranium (VI) concentrations in aqueous and organic solutions was used extensively. A summary table of final correlation equations, corresponding correlation coefficients and the average curve error (from test samples) is presented in Table 4.1.

5. EXPERIMENTAL RESULTS

5.1. Electrocell

The cell was first tested as a free-standing unit, to determine the optimum U(VI) reduction conditions as a function of differing flow rates and voltages for a solution of constant uranium (VI) and nitric acid concentrations. The data was then used to develop a mathematical equation which related the reduction

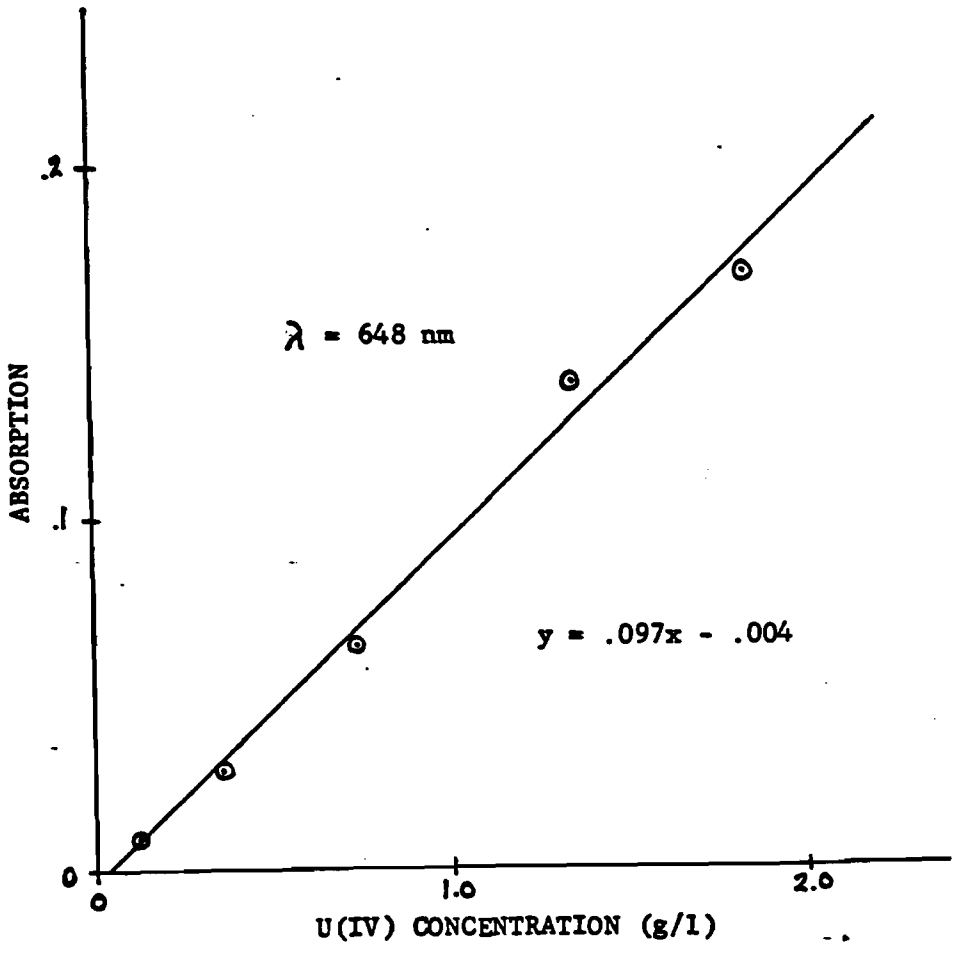


Figure 4.7 U(IV) Colorimetric Correlation - Organic

Table 4.1 Summary of Colorimetric Calibration Equations

Correl. Eqr. #	Uranium Valence	Phase	Acidity	Correlation(M)	Correlation(g/l)	Correl. Coeff.	Avg. Error	Standard Deviation
4.1	U(VI)	A	0	X=(Y+0051)/7.4641	X=(Y+.0051)x31.885	.99996	±1.69%	1.74%
4.2	U(VI)	A	1	X=(Y+.0091)/7.961	X=(Y+.0091)x29.878	.99992	±1.96	2.22%
4.3	U(VI)	A	3	X=(Y+.0031)/9.2851	X=(Y+.0031)x25.632	.99993	±1.98%	2.31%
4.4	U(VI)	O	—	X=(Y+.0071)/7.8381	X=Y+.0071)x30.370	.99921	±3.53%	3.09%
4.5	U(IV)	A	2.8*	X=(Y+.012)/27.6891	X=(Y+.012)x8.534	.99975	±3.40%	3.95%
4.6	U(IV)	O	—	X=(Y+.004)/23.111	X=(Y+.004)x10.298	.99478	±5.30%	5.72%

X = Uranium concentration
Y = Spec. absorption reading
*2.8 M Free Nitric Acid, .2 M hydrazine

capability of the cell to the U(VI) concentration in the stream, the flowrate and the voltage applied. The mathematical relation was used in the development of the computer program "ELECTRO", written to model the cell. During the testing of the cell, it was found that at very high currents (8 amps or larger) the hydrazine stabilizer was rapidly destroyed. Anodic destruction of hydrazine was previously reported [18][6].

5.1.1. Testing

The electroreduction cell was tested qualitatively by running a solution of uranyl nitrate through it with an arbitrary voltage applied. The solution was observed to change color from bright yellow, characteristic of uranium (VI), to green, which is characteristic of uranium (IV). This proved that the reduction cell was capable of reducing U(VI) to U(IV) without the need for a diaphragm. However, at the cell exit, foaming of the greenish liquid was observed, along with a return to its original yellow color. This reoxidation of the U(IV) was probably due to the oxygen evolved at the anode or by reaction with nitrite ions produced near the cathode [14]. The foaming can be explained either by this evolved oxygen or by the evolution of oxides of nitrogen [13]. The production of hydrogen may also have contributed to this foaming.

This initial test proved that the cell was capable of U(VI) reduction, but that the U(IV) produced was very unstable and reoxidized to U(VI) quickly in

the nitric acid medium. The holding reductant hydrazine was subsequently added to all solutions prior to reduction to prevent reoxidation. A concentration of 0.2 M hydrazine was used for all reduction tests [25][7]. No uranium (IV) reoxidation was observed in the presence of this concentration of hydrazine.

A series of uranium reductions were conducted using a constant uranyl nitrate solution of 0.157 M uranium (VI), 2.78 N free nitric acid, and 0.2 M hydrazine. The uranium concentration selected was arbitrary, but the nitric acid concentration was maintained at 2.78 N since it had been decided that all aqueous solutions used in the partitioning experiments would be at approximately that acidity. A literature search verified that most uranyl nitrate reduction and uranium partitioning experiments were performed at nitric acid acidities of 2.0 and 3.0 N [7][28][8][23]. Excess free nitric acid was necessary since it is consumed in the reduction of the uranyl ion. Also, the reduction rate of uranyl nitrate increases with increasing acidity [7].

The cell was tested by reducing this solution at different voltages and flow rates. The voltages were varied between 2.0 and 2.5 volts and the flow rates between 4 and 15 milliliters per minute. The voltages were somewhat difficult to control since an initial voltage supply setting that gave 2.5 volts would often decrease slightly as time increased (current was observed to increase

concurrently). Voltages of less than 2 volts were not used since very little color change was observed below that voltage and voltages greater than 2.5 volts were not included because gas evolution and foaming occurred above that potential.

After setting the flow rate and voltage, the cell was allowed to reach steady state, based on the solution's residence time in the cell (cell volume/flow rate). A sample was taken from the exit stream, the current was noted, the voltage was changed, and the cell was again allowed to reach steady state. The uranium (IV) concentrations of the samples were determined and the percent reduction for each sample was calculated along with the current efficiencies. Current efficiencies were determined by dividing the number of Faradays necessary (given 100% current efficiency) to produce the U(IV) content of the sample by the actual number of Faradays used (calculated with the known current and residence time for the sample).

The actual reductions ranged from 10.4% to 28.0%. The residence times ranged from 3.2 to 11.9 minutes and the current efficiencies were as high as 98.9% (at 2 volts) and as low as 7.9% (at 2.5 volts). The current efficiency of the cell appeared to decrease as the voltage increased, due to the increased electrical resistance, cell heating, and gas evolution which were observed at higher voltages [14]. Experimental data are given in Table 5.1

Table 5.1 Experimental Results

ELECTROLYTIC URANIUM REDUCTION

EXPERIMENT NO.	1	2	3	4	5	6	7	8
APPL. POT.,VOLTS	2.00	2.00	2.25	2.25	2.25	2.50	2.50	2.50
CURRENT,AMP	0.7	0.6	1.6	1.4	1.7	7.2	4.5	5.2
FLOW,ML/MIN	4	8	4	8	15	4	8	15
RES. TIME,MIN	11.9	6.0	11.9	6.0	3.2	11.9	6.0	3.2
U(VI) RED.,%	28.0	14.8	28.0	20.1	10.4	28.0	23.3	14.8
CURR. EFF.,%	80.9	98.9	35.6	57.3	44.2	7.9	20.7	21.4

CELL VOLUME: 47.6ML

CATHODE AREA: 92.7CM^2

INITIAL CONCENTRATIONS: U(VI) 0.157M

HNO_3 2.780N

N_2H_4 0.200M

5.1.2. Modeling

The extent to which the electrocell reduced U(VI) to U(IV) was found to be dependent on the flow rate, the initial U(VI) and nitric acid concentrations, the applied voltage, the cell holdup, cell volume and the electrode materials and surface areas [22]. To model the reduction of U(VI), these variables were grouped into a geometric factor (Ω) and an electrical factor (β). The change in U(VI) concentration (amount reduced) was then a function of the initial U(VI) concentration, the geometric factor (Ω) and the electrical factor (β) described by the following differential equation:

$$\frac{d\,[U(VI)]}{dt} = -\,[U(VI)] \times \Omega \times \beta$$

The electrocell was considered a plugged-flow reactor with little backmixing. At steady state the relationship between the concentrations of U(VI) in the entering and exiting streams was found by integrating the above rate equation for a batch reactor for a period of time t:

$$\ln \frac{[U(VI)]_{out}}{[U(VI)]_{in}} = -\beta\Omega t$$

$$[U(VI)]_{out} = [U(VI)]_{in} \exp(-\beta\,\Omega\,t)$$

where, t = residence time in seconds (= cell volume/flow rate)

Ω = geometric factor in cm^{-1} (= area of cathode/cell volume)

β = electrical factor in cm/sec (= function of voltage, electrode material, and hydrodynamics)

The volume of the cell and cathode area were 47.6 cm^3 and 92.7 cm^2 respectively. The β factor was then calculated by solving the integrated rate equation, using the experimentally determined uranium concentrations. A list of the values obtained for each point is presented in Table 5.2.

Inspection of the beta values obtained showed that they were not constant but varied with the voltage and the flow rate (presumably because the flow rate affects the cell turbulence and thus the effective "free" cathode area). A multivariable linear regression program was used to derive an equation to model this factor. Correlations were made for the flow rate, the square root of the flow rate, and the natural log of the flow rate. The best results were obtained using the natural log of the flow rate. This gave a coefficient of determination (R^2) of 0.79 and the following equation for β:

$$\beta = 0.019\,(V) + 0.00583\,(\ln FR) - 0.0364$$

The results are presented in Table 5.2. Using this function for the β factor in the original U(VI) reduction equation, along with the geometric factor and residence time, gives:

$$[U(VI)]_{out} = [U(VI)]_{in} \exp - \frac{92.7}{FR}(0.019\ V + 0.00583\ (\ln FR) - 0.0364)$$

The equation was used to generate the uranium conversion efficiencies for each of the data points used to develop the equation, so that the fit of the model to the actual data could be observed. A comparison of calculated and experimental data is presented in Figure 5.1.

Table 5.2 Cell Modeling Data

Variables: X=Voltage
Y=Function of flow rate
Z=β

Y Variable	Coefficient of Determination (R^2)	a	b	c	Equation (β model)
FlowRate	.78	-.0295	.0185	.00068	
√FlowRate	.76	-.0304	.0173	.0036	
*ln(Flow Rate)	.79	-.0364	.0190	.00583	β=.019(V)+.00538(lnFR)-.0364

Cell Reduction Equation: $[U^6]_{out}=[U^6]_{in}\exp-\frac{(92.7)(.019(V)+.0058^3(\ln FR)-.0364)}{(FR)}$

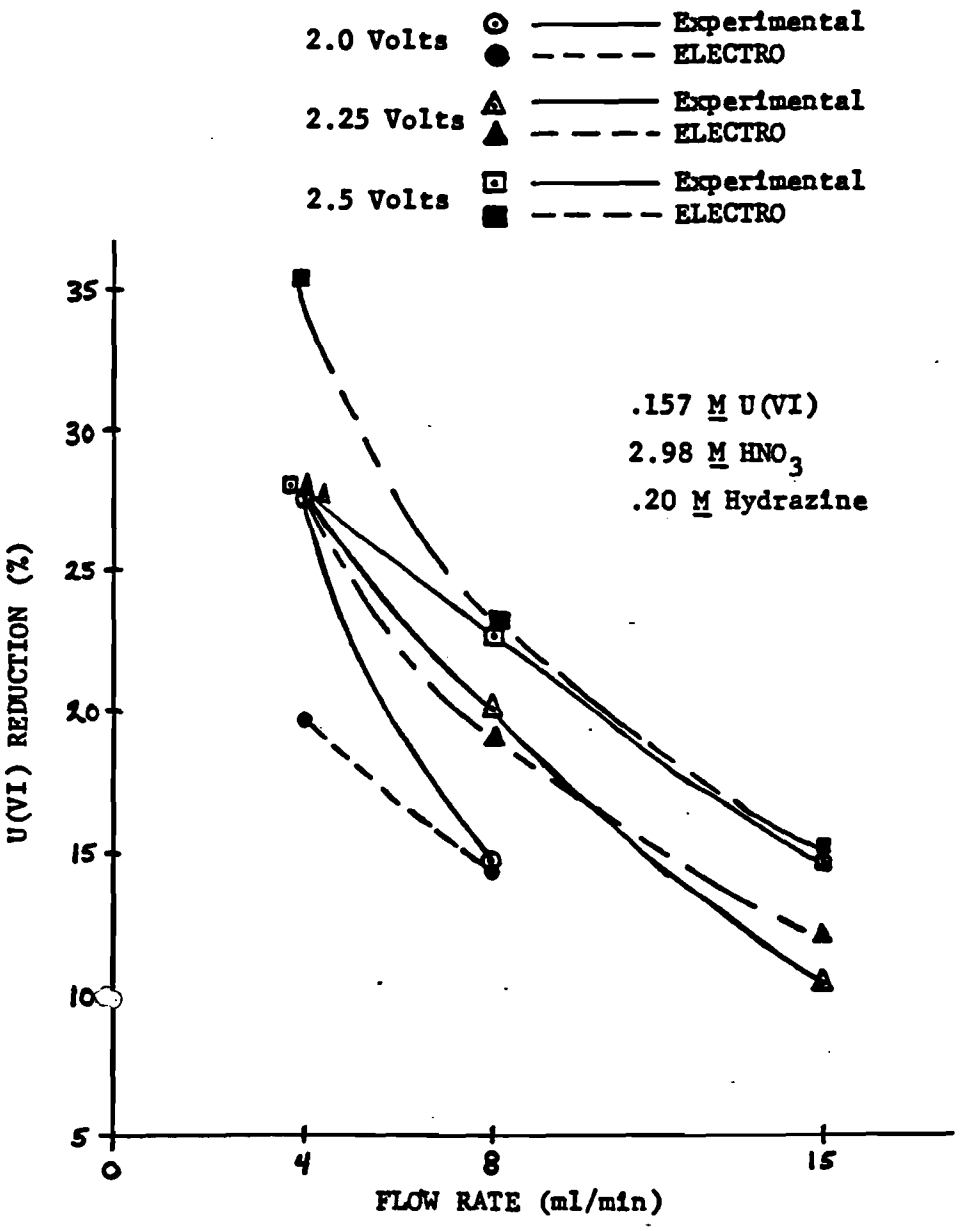


Figure 5.1 ELECTRO Versus Experimental Electrocell Results

The equation modeled the cell for the reduction of U(VI) alone, but to model a stream in which plutonium was present the direct reduction of Pu(IV) had to be accounted for as well as the reduction of Pu(IV) by U(IV). Petrich, et. al., gave the following equations for plutonium reduction [12]:

$$\frac{d\,[Pu(IV)]}{dt} = -\beta\Omega[Pu(IV)]$$

$$\frac{d\,[Pu(IV)]}{dt} = \frac{-\delta\,[Pu(IV)]\,[U(IV)]}{[HNO_3]^{-2}}$$

These two equations, along with the equation for direct U(IV) reduction, were included in the program ELECTRO, for an electrolytic reduction cell.

Solution of the first equation was accomplished by simple integration, similar to the previous U(VI) reduction equation. The Pu(IV) reduction by U(IV) equation was more complicated however, since it contained more than one variable. A finite elements method was used, whereby the cell was divided into a number of subdivisions. The residence time of the liquid in a subdivision is very short, so that the concentrations of U(VI), Pu(IV), U(IV) and HNO_3 are assumed to remain constant within the subdivision and are set to equal the concentrations at the entry to the subdivision. A complete material balance is then performed to obtain the concentration in the stream leaving the subdivision.

ELECTRO was written to allow the user to set the number of subdivisions between 1 and 99, depending on the level of accuracy desired. A listing of the program, and sample input and output files are presented in Appendix B.

To test how the results change with the number of sub-cells, identical inputs were run and the number of divisions was varied between 99 and 3. It was found that the percent reduction expected did not decrease significantly in accuracy until a very small number of subcells was used. Even for just three sub-cells the expected reduction (8.3%) was only 3.1% smaller than the reduction calculated for 99 sub-cells (12.0%). The program output for these runs is presented in Table B.3.

5.1.3. ELECTRO Results

The results calculated with ELECTRO were not entirely successful. Of the three simulated partitioning experiments for which the reduction portion of the experiment was modeled, ELECTRO underestimated and overestimated the actual reductions found. The discrepancies observed may be explained by certain factors which were not taken into account by ELECTRO, primarily the reoxidation of U(IV) and side reactions. Also, a constant value was used for β, rather than a variable dependent on voltage and flow rate.

In the first simulated partitioning experiment the voltage supply was maintained at 3.5 volts and the flow rate of the aqueous streams was 15.7 ml/min. At these settings the ELECTRO model estimated a 24% reduction

rate. The actual experimental reduction, obtained by comparing the U(VI) aqueous concentrations before and after the cell, was found to be 35%. In the second simulated partitioning experiment the ELECTRO program projected a 27% reduction compared to the actual 30% reduction calculated. In the third experiment, at 3 volts and an aqueous flow rate of 5.7 ml/min, the ELECTRO program predicted 39% reduction. The actual reduction for these conditions was 10%.

In the first simulated partitioning run, in which the actual reduction was underestimated by about 10%, it should be noted that the flow input to the electrocell was somewhat erratic due to booster pump problems. This is significant since higher reduction rates have been found for agitated rather than static solutions [14]. In addition, the higher flow rate in this experiment, as opposed to the second and third partitioning runs, may have contributed to more turbulent flow in the cell.

In the third simulated partitioning experiment the cell produced a reduction rate of 10%, compared with a predicted reduction rate of 39%. The second and third simulated partitioning experiments were run under almost identical electrocell conditions (2.5 volts and 5.7 ml/min versus 3.0 volts and 5.7 ml/min, respectively), but gave drastically different reduction rates (30% versus 10%, respectively). This indicates that the discrepancy seen in the third experiment was due to reduced cell performance rather than problems with ELECTRO's modeling of the cell.

Investigation of the cell's internals verified that the titanium cathode had a dull grayish layer on it which could be scraped off. The use of the cell as an "oxidation" cell for hydrazine destruction preceding the third experiment may have contributed to this buildup. Other possibilities include the presence of iron, aluminum, or TBP in the aqueous stream.

5.1.4. Hydrazine Destruction

In the process of reducing a solution already containing U(IV) and effectively stabilized with hydrazine it was found that increasing the current beyond about 8 amps resulted in foaming and active gas evolution and 100% reoxidation of U(IV) in the solution as determined by colorimetric analysis. This suggested that the hydrazine stabilizer had been fully destroyed. Studies have shown that the primary hydrazine destruction method is by reaction with oxygen generated at the anode [13]. Also, direct electrolytic oxidation of hydrazine may occur at the anode by the equation [6]:

$$N_2H_5 \longrightarrow N_2 + 5\,H^+ + 5e^-$$

The electrocell was used several times as an oxidation cell for the destruction of hydrazine and reoxidation of U(IV). This process was very effective but relatively slow due to the small throughput of the cell. The use of the cell as an "electro-oxidation" unit was achieved by setting the current between about 8 to 12 amps. Care was taken to keep the cell area well ventilated since high current densities at the cathode can produce hydrogen [18]. In the direct anodic oxidation process hydrazine is converted into molecular nitrogen and hydrogen.

5.2. Centrifugal Contactor

5.2.1. Stage vs. Cascade Testing

An extraction run was made with uranium to test the operation of the contactor. Satisfactory performance was indicated from the exit stream analyses, but compared with the SEPHIS model the stagewise concentration profiles revealed unsatisfactory performance in several of the stages due to internal flooding. Tests were run on the individual stages of the contactor by introducing 0.1 N nitric acid and 30% TBP solutions directly to that stage and observing its exit streams. Hydraulic performance was considered acceptable if both exit streams had less than 1% phase contamination [5].

These tests showed flooding in either the aqueous or the organic exit streams was occurring in four stages. It was determined that the individual rotors were the cause of the problems, since movement of a "bad" rotor to a previously satisfactory stage position in the contactor housing produced unsatisfactory results. Each of the four rotor-bearing assemblies was removed and carefully cleaned. This had no effect so the rotors were removed from each of the bearing assemblies and attempts were made to reposition the rotors back onto their bearing assemblies. This was successful for only one of the four rotors.

Attempts to replace the defective rotors with newly machined (never used) rotors was successful in two of the remaining three rotors. The defective rotors showed indications of wear and tear and may have contained mechanical

defects or internal obstructions in the rotor channels or apertures. Further efforts to clean, reposition, or replace the remaining defective rotor were unsuccessful in all stages of the contactor housing. The cause of this may have been a slightly bent or warped bearing shaft. This could have allowed one phase to enter in the wrong locations, due to the tilted position of the rotor.

Because all efforts to make the final rotor-bearing assembly operable were unsuccessful and no new bearing assemblies were available, it was necessary to scale the contactor down from an eight-stage to a seven-stage unit. To accomplish this, the working rotors were shifted so that the original aqueous entrance and organic exit stage (stage one) was skipped completely. The aqueous entrance and organic exit now occurred in the stage previously termed stage two. Stage two was renamed stage one and all other stages were numbered correspondingly up to stage seven (the aqueous exit and organic entrance point).

Subsequent testing of the contactor gave satisfactory results for most conditions, but some flow surging between stages was observed. Previous studies of the 2-cm ANL centrifugal contactor have confirmed that the interstage flow can fluctuate significantly due to the cyclic variation of the dispersion level in the mixing zone [5]. Initial development work at ANL assumed that whatever results were observed in single-stage operation would also be observed in multistage operation. Measurements, however, indicated

that in multistage operation there was a loss of maximum throughput and extraction efficiency due to flow surging [16][17]. The multistage capacity, at any O/A ratio, has been determined at ANL to be 80% of that for single-stage operation of the 2-cm contactor. Also, at low flow rates a reduction in the O/A operating range has been attributed to the variation of flow rate with time [5].

5.2.2. Hydraulic Operability Range

To determine the operability range of the contactor with respect to its rotational speed, an experiment was performed in which the total throughput of the contactor was varied for a given contactor speed. The rotational speed of the contactor was then adjusted and the experiment repeated. A consistent O/A ratio of one was maintained throughout the test. The aqueous and organic test streams were 3 N nitric acid and 30% TBP solutions, respectively. The total throughput was varied between 8 and 50 ml/min and the contactor speed was varied between 4000 and 7400 rpm. The speed of the turning rotors was determined with a Power Instruments model C-891 stroboscope. The same 1% phase contamination criterion used for single stage tests was used to determine contactor performance.

Based on the test results which showed the best operability between 5800 and 6600 rpm, all successive experiments were done with a contactor speed of 6200 rpm. The results of the test are presented in Figure 5.2.(a). The average total volume of liquid in the seven-stage contactor at 6200 rpms was 210 ml.

Seven-stage Contactor

Aqueous: 3.0 N HNO_3

Organic: 30% TBP, 70% Norpar

O/A Ratio: 1

● Satisfactory Performance (continuous exit phases)

○ Unsatisfactory Performance (phase contamination)

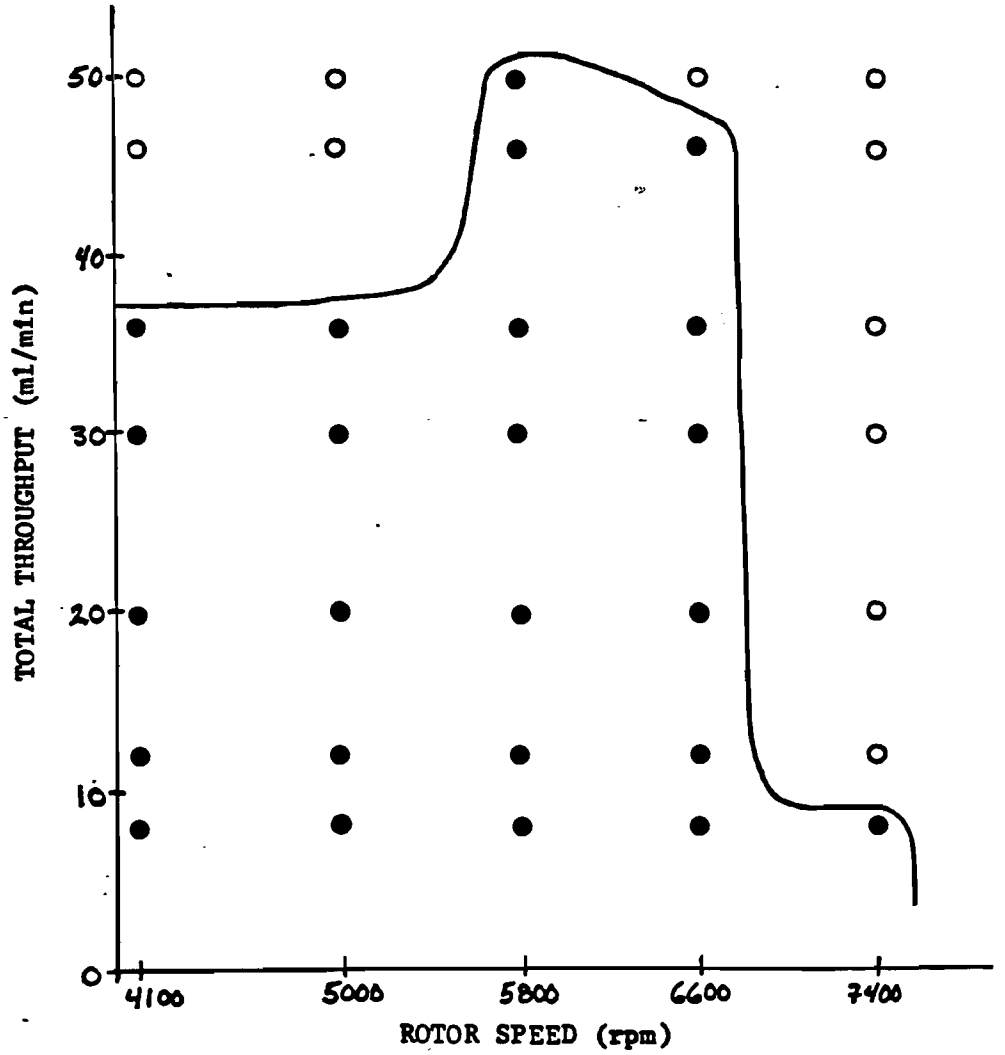


Figure 5.2.a Contactor Hydraulic Performance

Next the hydraulic operability range of the contactor was tested for a wide range of total throughputs and O/A ratios. This was accomplished by holding the total throughput constant and varying the O/A ratio from 8/1 to 1/8. The range of throughputs tested was from 10 to 36 ml/min (limited by the available pumps). The aqueous and organic streams were 3 N nitric acid and 30% TBP solution, respectively. The contactor speed was maintained at 6200 rpm and 1% exit phase contamination was again adopted as the limit for satisfactory performance.

The results for each throughput and O/A ratio were characterized as either aqueous-or-organic continuous (indicating which phase was dispersed in the other "continuous" phase) and as either satisfactory or unsatisfactory (indicating whether phase contamination was present or not). Identification of the continuous phase was accomplished by observing the appearance of the exiting streams. Normally, the phase which has been the continuous phase in the mixing zone is cloudy after separation and the previously dispersed phase is clear after separation. It should be noted that the dispersion of the aqueous phase in the organic occurs more easily than does the reverse [5]. The results of these tests are presented in Figure 5.2.(b).

As is apparent in Figure 5.2. (b), there were two distinct regions of satisfactory performance. The first satisfactory region was when the organic phase was continuous. That range encompassed virtually all combinations of throughput, from 10 to 36 ml/min, and O/A ratios from one to four; beyond an

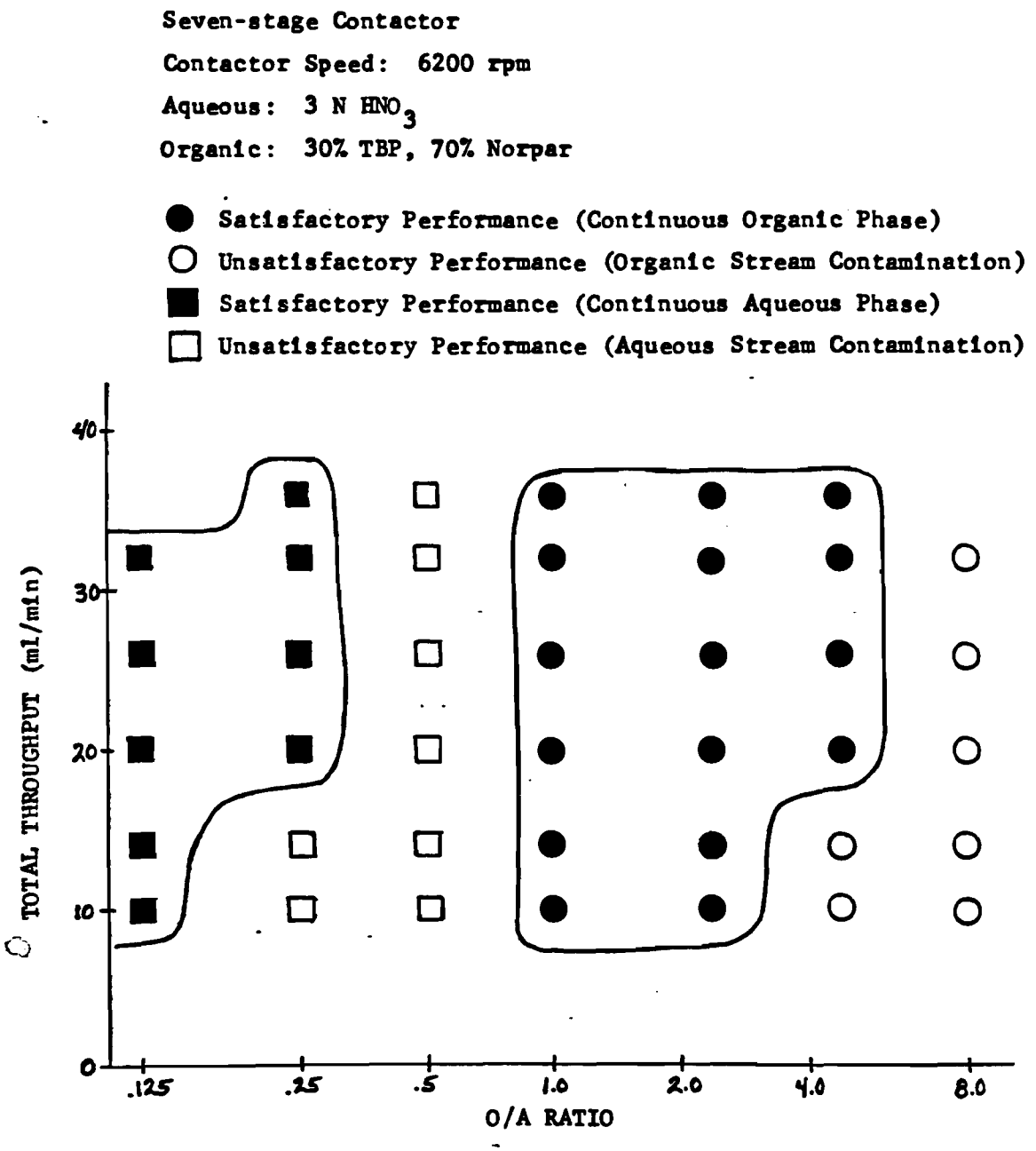


Figure 5.2.b Contactor Hydraulic Performance

O/A ratio of about four there was flooding of the organic phase into the aqueous phase. The second region of satisfactory performance occurred for O/A ratios of 1/4 or less and total throughputs between 14 and 36 ml/min. The area of unsatisfactory performance between the two regions, roughly covering the O/A ratios between 0.25 and 1.0 (except at lower flow rates), is called the organic-to-aqueous continuous phase inversion region [5].

With an initial organic-continuous state, as the O/A ratio falls below one there is a high resistance to phase inversion from organic-continuous to aqueous-continuous in the couette mixing zone. Instead of inverting the dispersion, the aqueous phase backs up in the annular mixing zone until some of it comes out with the organic exit stream. If the O/A ratio becomes low enough (at around 1/4 in this test), the phase inversion to aqueous-continuous can take place. By contrast the aqueous-to-organic continuous phase inversion occurs easily [5].

The resistance to phase inversion from organic- to aqueous-continuous causes the backup of liquid in the interstage feed lines. Just before a phase inversion occurs, the interstage back-pressure may become so high that liquid is forced out the stage purge-line just above the aqueous exit port [5]. This effect was observed many times during this study.

5.2.3. Contactor Plus Electrocell Hydraulics

The electrocell was placed in-line between stage one and stage two by replacing the aqueous interstage feed line with tygon tubing from the stage one exit to the electrocell entrance and from the electrocell exit to the stage two entrance. To test the hydraulics of the system, the aqueous phase was pumped into stage one and allowed to flow from stage one into the electrocell. The head developed by the contactor was not enough to overcome the pressure drop through the cell and backup of the aqueous into the organic exit stream occurred in stage one.

To overcome the pressure drop of the cell, an interstage pump was added. It was of identical size and design as the aqueous and organic entrance pumps. This pump was placed between the stage one exit port and the entrance to the electrocell. After synchronizing the flow rates of the booster pump and the original aqueous pump the booster pump was more than able to overcome the cell pressure drop and to pump liquid into stage two.

To ensure that the booster pump would not back up aqueous from stage one (if its pumping rate was too low) or create a suction at the stage one exit port (if its pumping rate was too high) a modification was made to the stage one exit. A T-type connector was added and a glass tube, partially filled with the original aqueous solution, was attached to the connector. This tube ensured that flow variations in the contactor would not seriously affect

operation of the system. The liquid level in the glass tube indicated whether the booster pump operated at the proper rate. Hydraulic tests (total throughput versus the O/A ratio) were repeated for the modified system; the satisfactory operability regions were identical to those obtained for the contactor alone.

5.3. Experimental vs. SEPHIS Results

Extraction and stripping runs were made to verify that the contactor was operating satisfactorily, and that the SEPHIS Mod 4 code could model it. The SEPHIS code modeled both experiments very accurately. Next, a simulated partitioning experiment, in which the U(VI) in an aqueous stripping stream was reduced in-line by the electrocell, was conducted. The results of this run indicated that the actual U(IV) concentrations dropped off more rapidly than was estimated by SEPHIS. Suspecting excessive hydrazine was being consumed in the cell, a second simulated partitioning run was made with twice as much hydrazine. The U(IV) reoxidation problem persisted, so a third run was made under an inert argon gas blanket. The results of this experiment were much more satisfactory.

Small variations in the experimental data were observed which may have been caused by refluxing of nitric acid (varying the distribution coefficients), temperature gradients, the possibility that the unit was not quite at steady-state when the stages were sampled, and the uneven withdrawal of interstage samples. The experimental conditions for this series of runs are summarized in Table 5.3. The SEPHIS outputs are in Appendix B.

Table 5.3 Solvent Extraction - Summary of Experimental Results

EXPERIMENT TYPE	FLOW RATE AQUEOUS ML/MIN	ORGANIC ML/MIN	AQUEOUS FEED U(VI) G/L	HNO3 N	N2H4 M	ORGANIC FEED U(VI) G/L	HNO3 N	POTENTIAL VOLTS	CURRENT AMPS	U(VI) REDUCED %	GAS BLANKET
EXTRACTION	12.2	12.1	98.6	0.10	0.00	3.4	0.04	0.0	0.0	0	AIR
STRIPPING	15.7	13.0	0.0	0.01	0.00	84.8	0.08	0.0	0.0	0	AIR
SIM.PART.#1	15.7	13.0	0.0	2.80	0.25	85.4	0.20	3.5	6.3	35	AIR
SIM.PART.#2	5.7	17.5	0.0	2.55	0.50	82.7	0.05	2.4	4.9	30	AIR
SIM.PART.#3	5.7	17.1	0.0	2.55	0.53	88.1	0.04	3.0	5.4	10	ARGON

5.3.1. Extraction Experiments

Initially, several test extraction runs were made. In both cases, however, the uranium in the aqueous stream was extracted almost entirely by the second stage. This left very little to compare with SEPHIS for the remaining stages, since the uranium was at practically undetectable levels. To spread the uranium extraction over the whole cascade, adjustments had to be made in the aqueous stream to render the extraction less efficient. Scoping runs with SEPHIS were made to determine what parameters should be used.

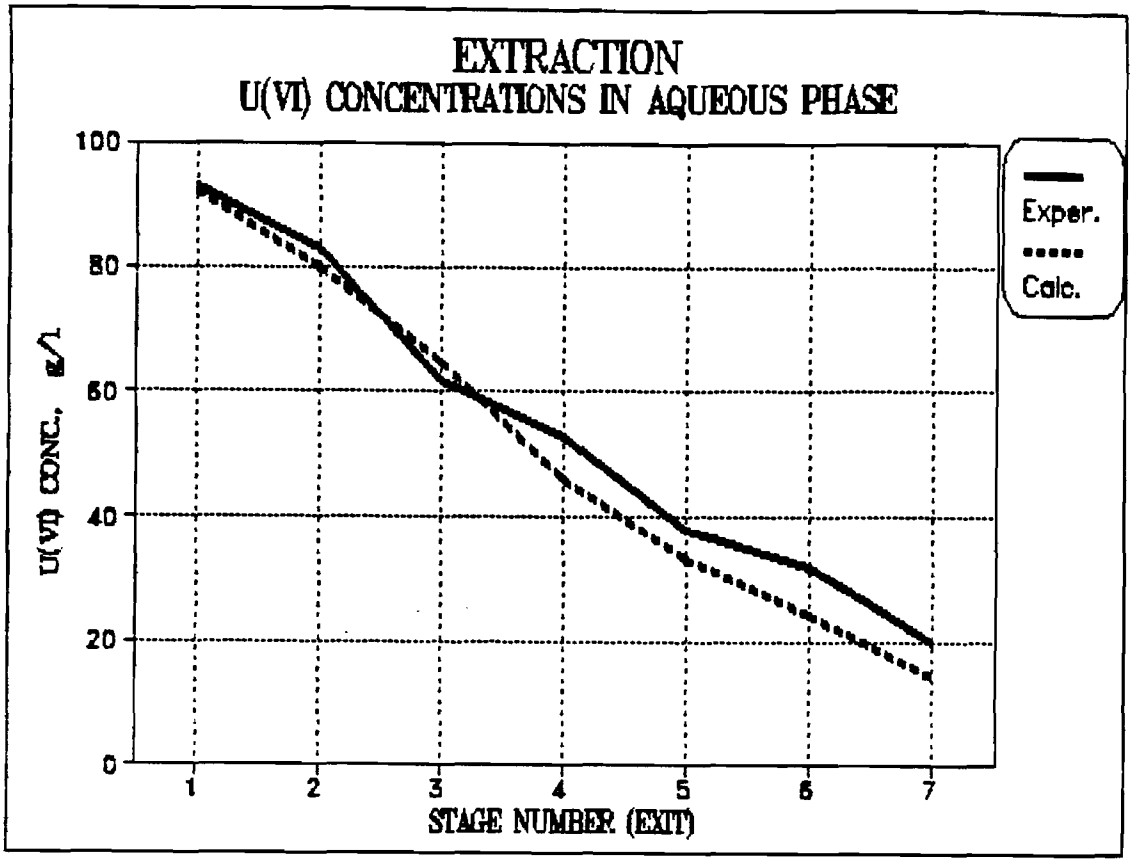
The extraction run parameters, based on the expected results from the modified SEPHIS Mod 4 program, were as follows:

Aqueous Stream: 98.6 g/l [U(VI)], 0.1 N [HNO_3], flow = 12.2 ml/min

Organic Stream: 3.4 g/l [U(VI)], 0.04 N [HNO_3], flow = 21.1 ml/min

(The presence of trace uranium and nitric acid in the organic solution is due to its previous use in batch extractions.)

The comparison of SEPHIS Mod 4's estimates and the actual experimental results for the aqueous and organic streams are presented in Figure 5.3. The results showed that the contactor was not encountering any stagewise discontinuities or rotor problems and that SEPHIS could model the process effectively.



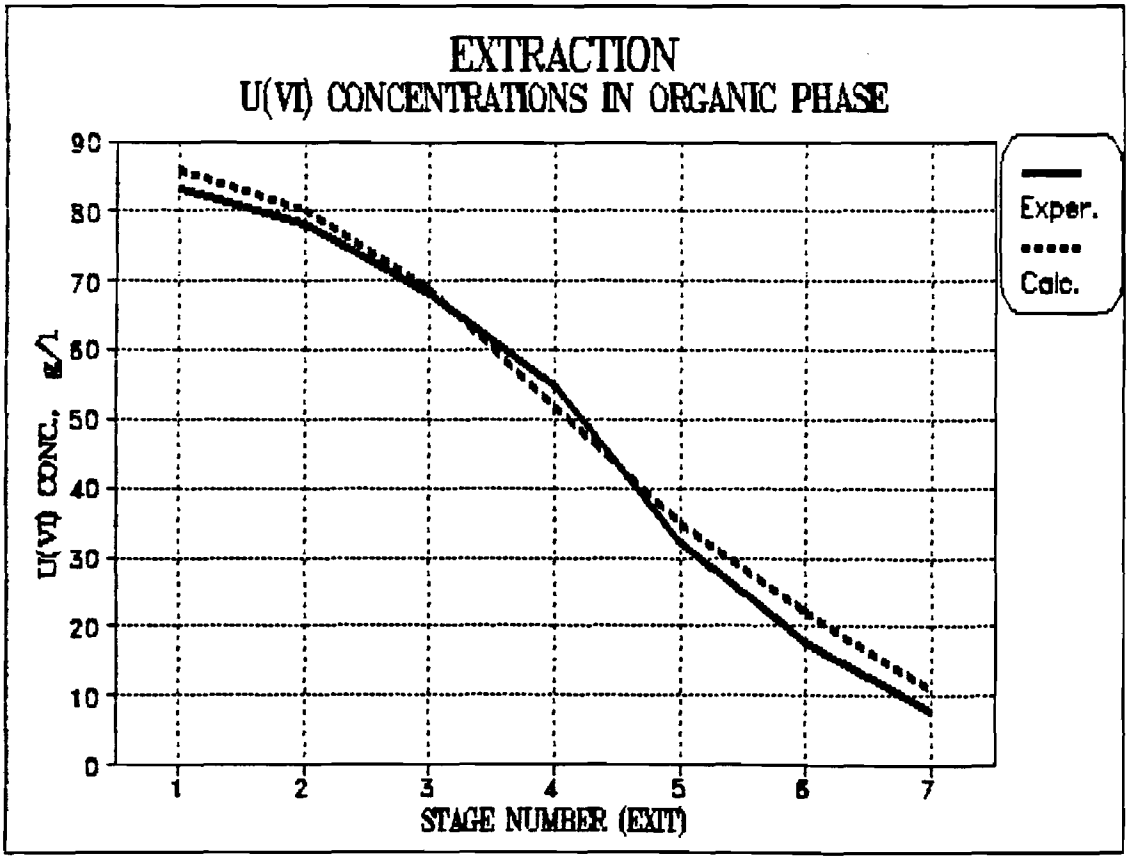


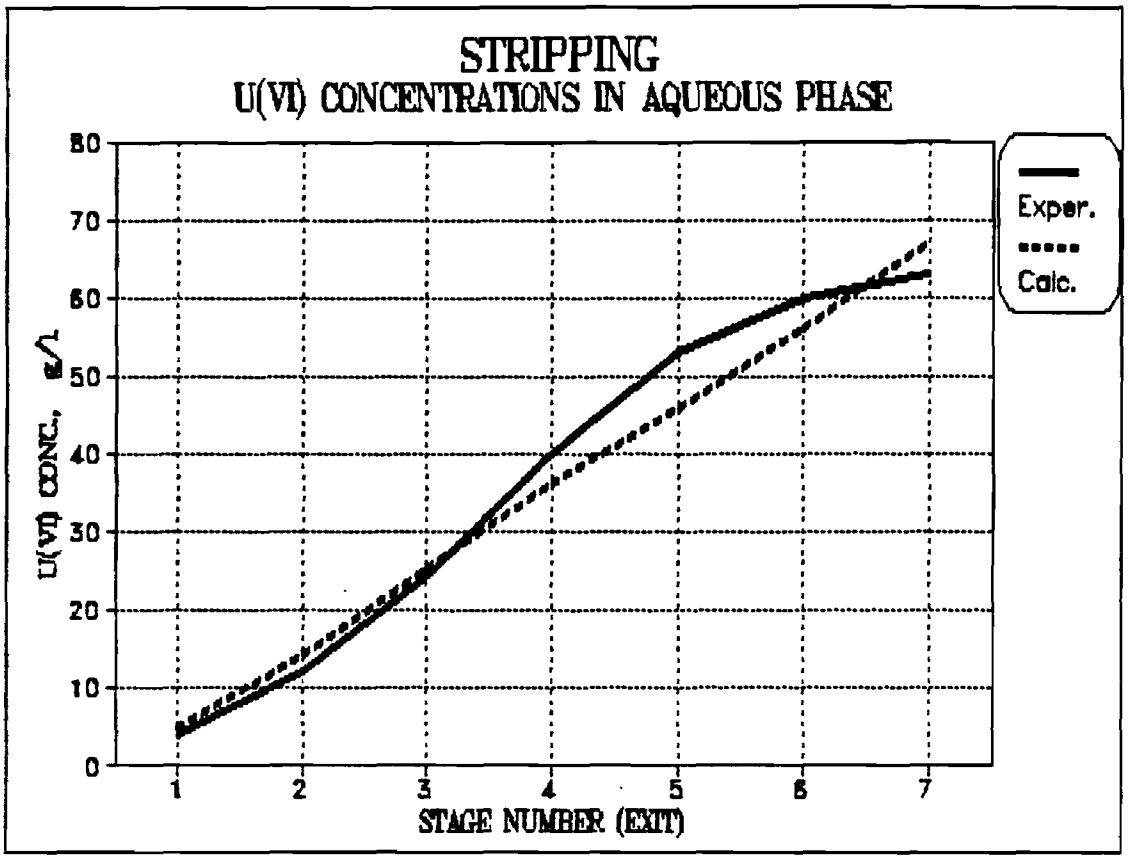
Figure 5.3 Extraction Experiment U(VI) Concentration Profiles

5.3.2. Stripping Experiment

To further verify the correct operation of the contactor and validate the SEPHIS Mod 4 program, a stripping experiment was conducted. The loaded organic extract from the test extraction experiment was used as the organic feed stream. It contained 84.8 g/l of uranium (VI) and was approximately 0.08 N in nitric acid. The aqueous stream was 0.01 N nitric acid and the O/A ratio was 0.83 (15.7 ml/min aqueous, 13.0 ml/min organic). A comparison of the calculated and experimental results for the aqueous and organic streams are plotted in Figure 5.4.

After the stripping test was begun, samples of the aqueous and organic exit streams were taken every five minutes for 35 minutes. The concentrations of U(VI) in both the aqueous and organic samples did not change after 20 minutes. Therefore, it was assumed that the system as a whole had reached steady state after 20 minutes. Individual stage samples were taken after 35 minutes.

It should be noted that this stripping test was conducted with the electrocell in place, as described in section 5.2.3., between stages one and two. The power was not turned on so the cell acted only as a delay line. The extremely close modeling of the test by SEPHIS again proved that the presence of the cell did not hydraulically or otherwise influence the operation of the contactor.



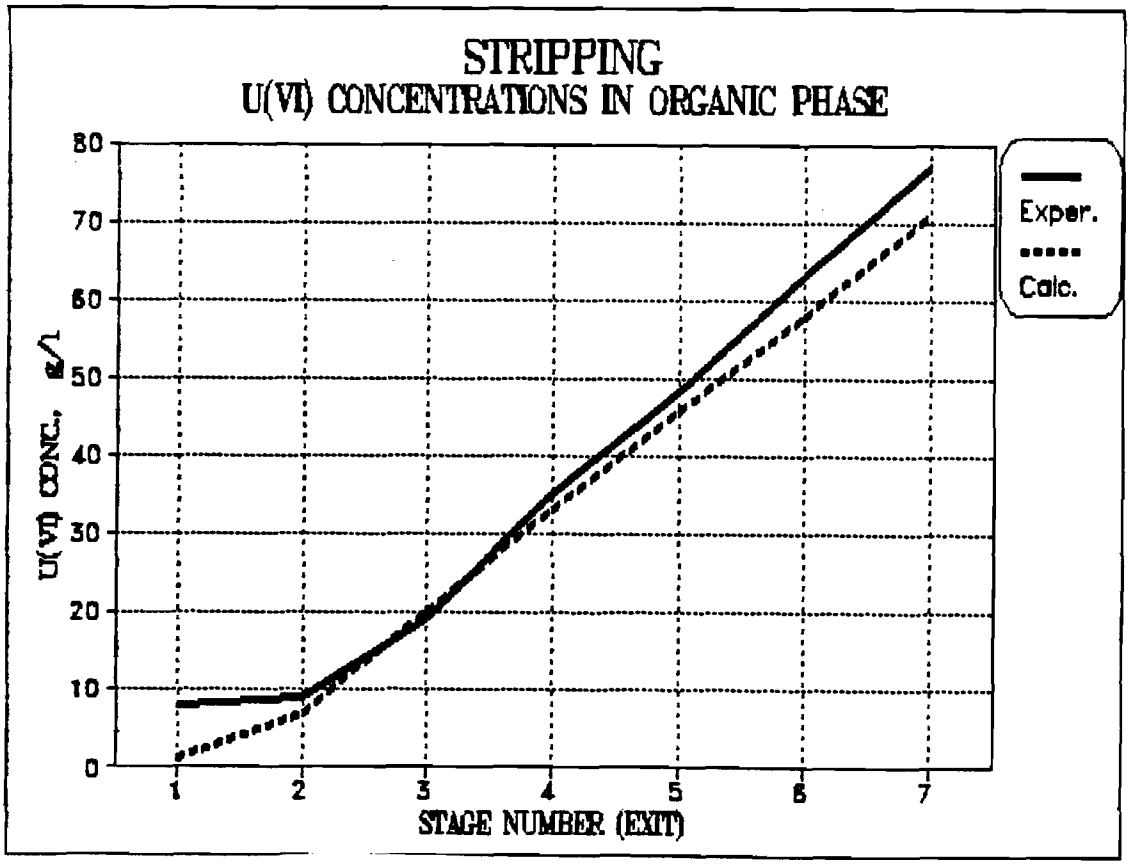


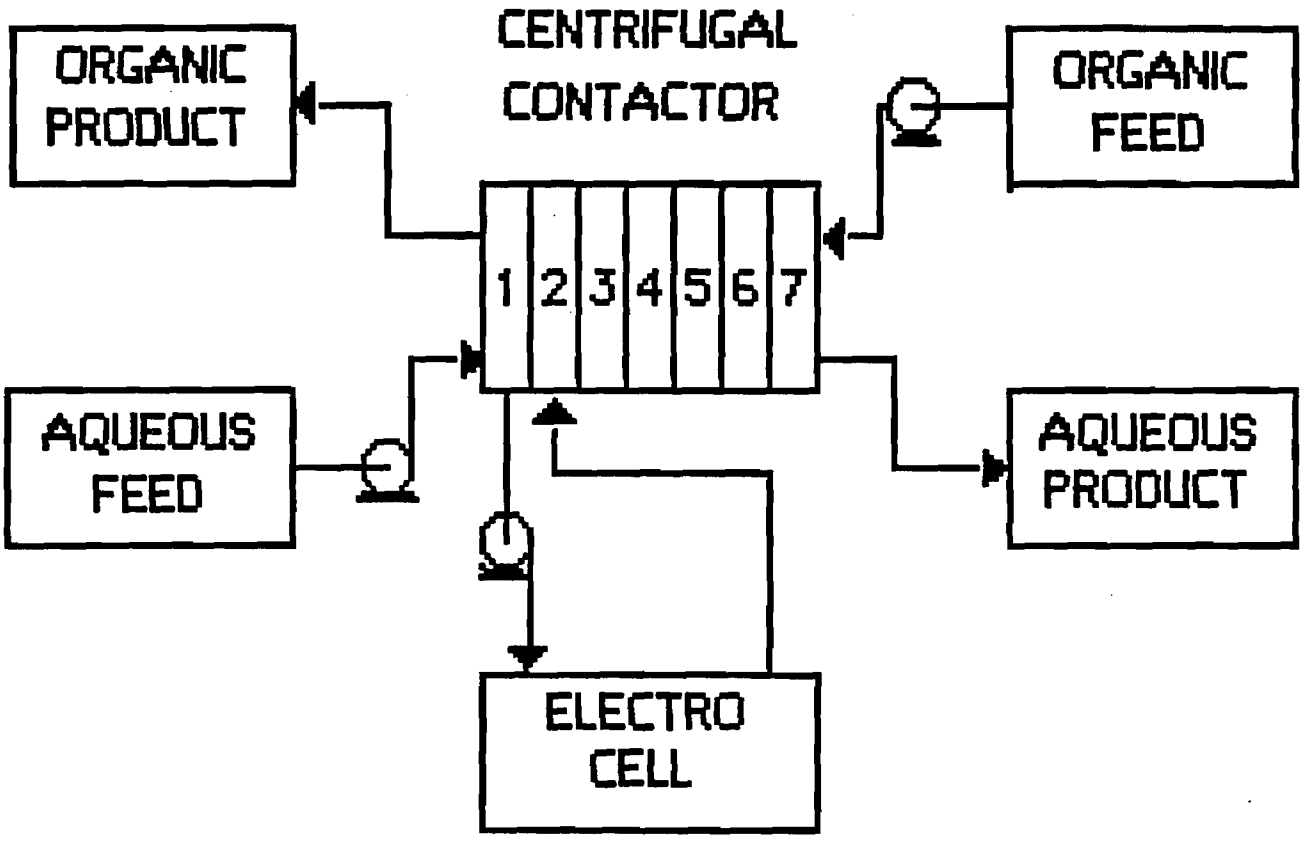
Figure 5.4 Stripping Experiment - U(VI) Concentration Profiles

5.3.3. Simulated Partitioning Experiments

The experiments up to this point showed that the contactor was operating correctly; the electrocell was operational and capable of at least 28% U(VI) reduction, that the addition of the booster pump did not interfere with the hydraulics or operation of the contactor and that SEPHIS Mod 4 calculated concentration profiles quite accurately. The modification of SEPHIS to incorporate modeling of the electrocell had not yet been completed. As an interim measure, the SEPHIS Mod 4 code was modified to model the reduction of a fixed amount of U(VI) to U(IV), by removing the aqueous stream between stages one and two, reducing a portion of it and sending it back to stage two. The arrangement of the equipment is shown in Figure 5.5.

Simulated Partitioning Experiment #1:

The purpose of the simulated partitioning experiment was to validate the modified version of SEPHIS for streams containing uranium (IV) and uranium (VI). The aqueous stream had a nitric acid concentration of 2.8 N, a hydrazine content of 0.25 M and flowed at a rate of 15.7 ml/min. The organic stream was maintained at a flow rate of 13.0 ml/min and had U(VI) and nitric acid concentrations of 85.4 g/l and 0.1 N, respectively. The electrocell voltage was adjusted to 3.5 volts resulting in a current of 6.3 amps.



ELECTROREDUCTION PARTITIONING SCHEME

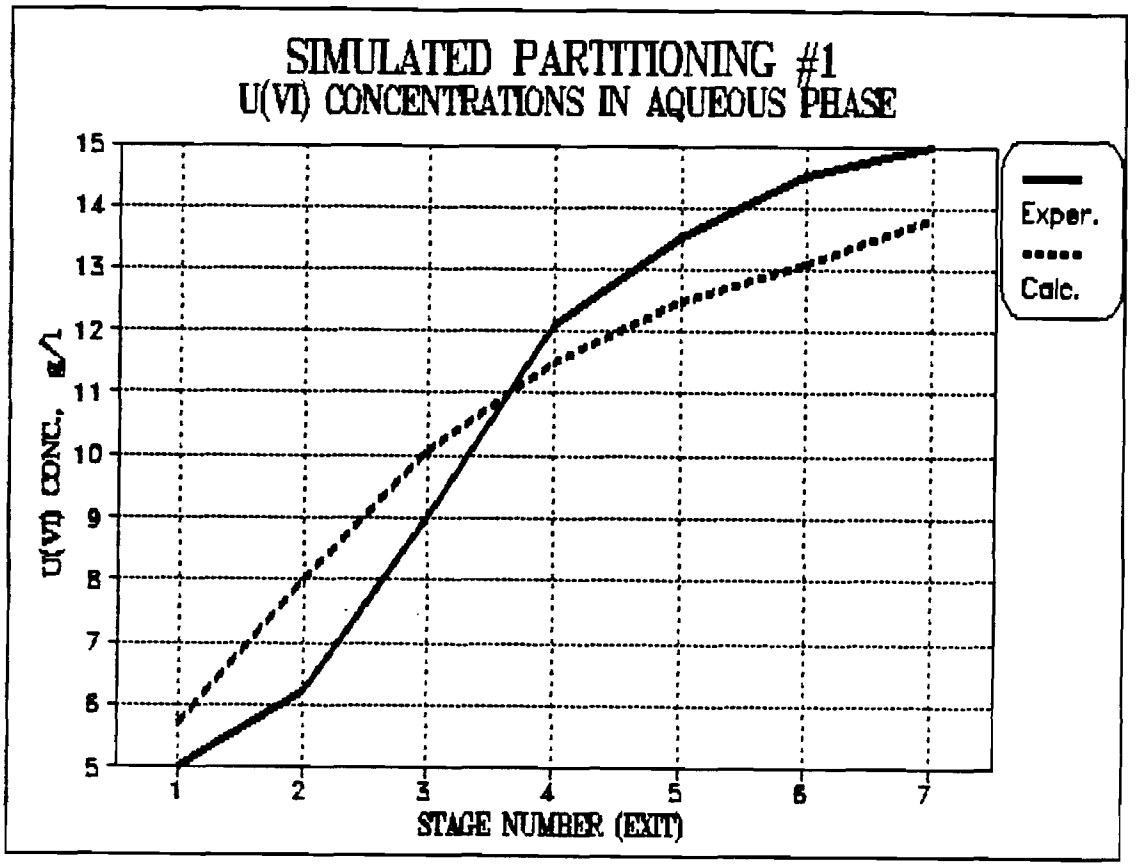
Figure 5.5 Electroreduction Partitioning Scheme

Analyses of aqueous exit streams, taken at five minute intervals, showed that equilibrium had been reached for this experiment after 25 minutes. Stage samples were taken after 45 minutes. Analyses of the U(VI) content in the aqueous stream just before entrance to the electrocell and the U(VI)/U(IV) content just after the cell provided an estimate of the percent reduction achieved. The actual U(VI) reduction produced by the cell was approximately 35%. This was the value used as input for percent reduction in the modified SEPHIS program. The aqueous stream temperature was monitored at the cell exit and found to have increased from 25 degrees to 27 degrees Celsius.

A comparison of SEPHIS estimates with the actual experimental results are presented in Figures 5.6 and 5.7.

The graphs show that the aqueous and organic U(VI) concentrations were modeled relatively well, but there were large differences between the expected and observed concentrations of U(IV) in the aqueous and the organic streams. It is obvious from Figure 5.7 that, although the amounts of U(IV) exiting the cell agree, the experimental U(IV) values drop off more rapidly than SEPHIS predicted.

The explanation for this was that either the U(IV) in the actual experiment was extracted more readily than expected into the TBP (meaning that the U(IV) distribution coefficient in the modified SEPHIS program was in error) or that the U(IV) present was being reoxidized to U(VI). Inspection of



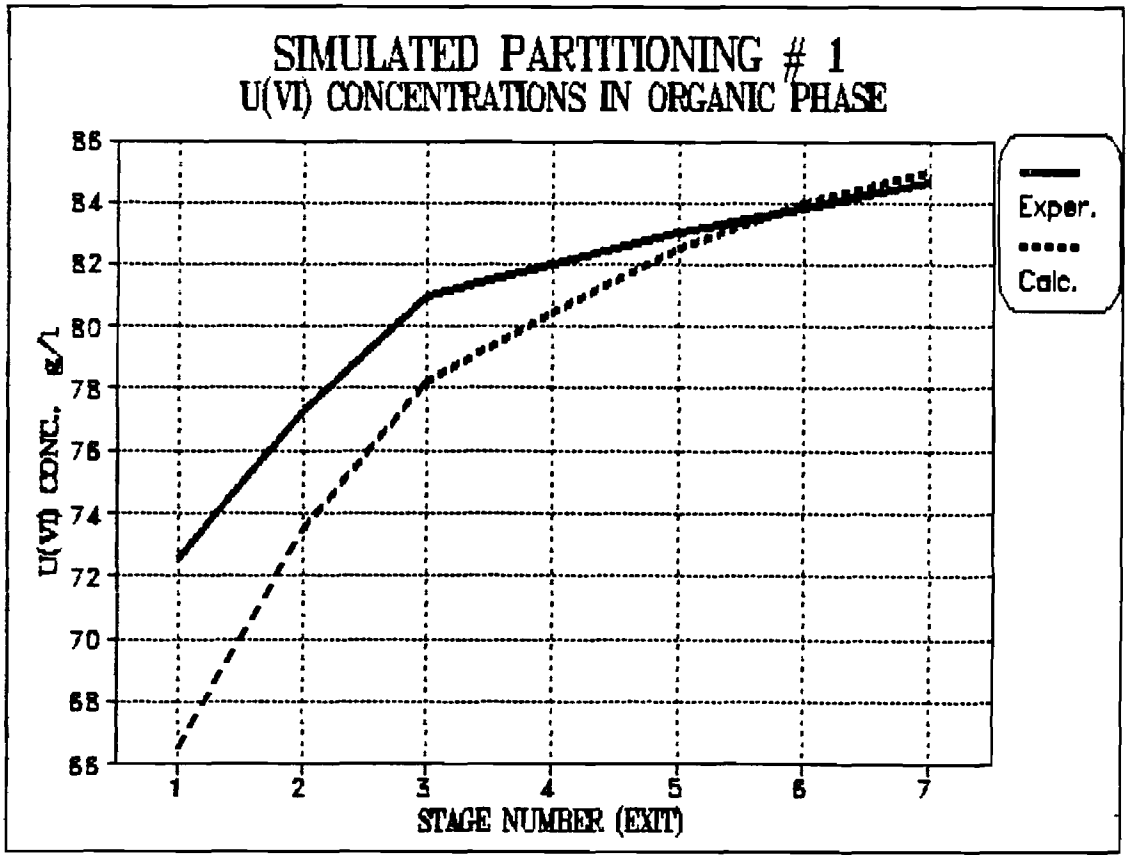
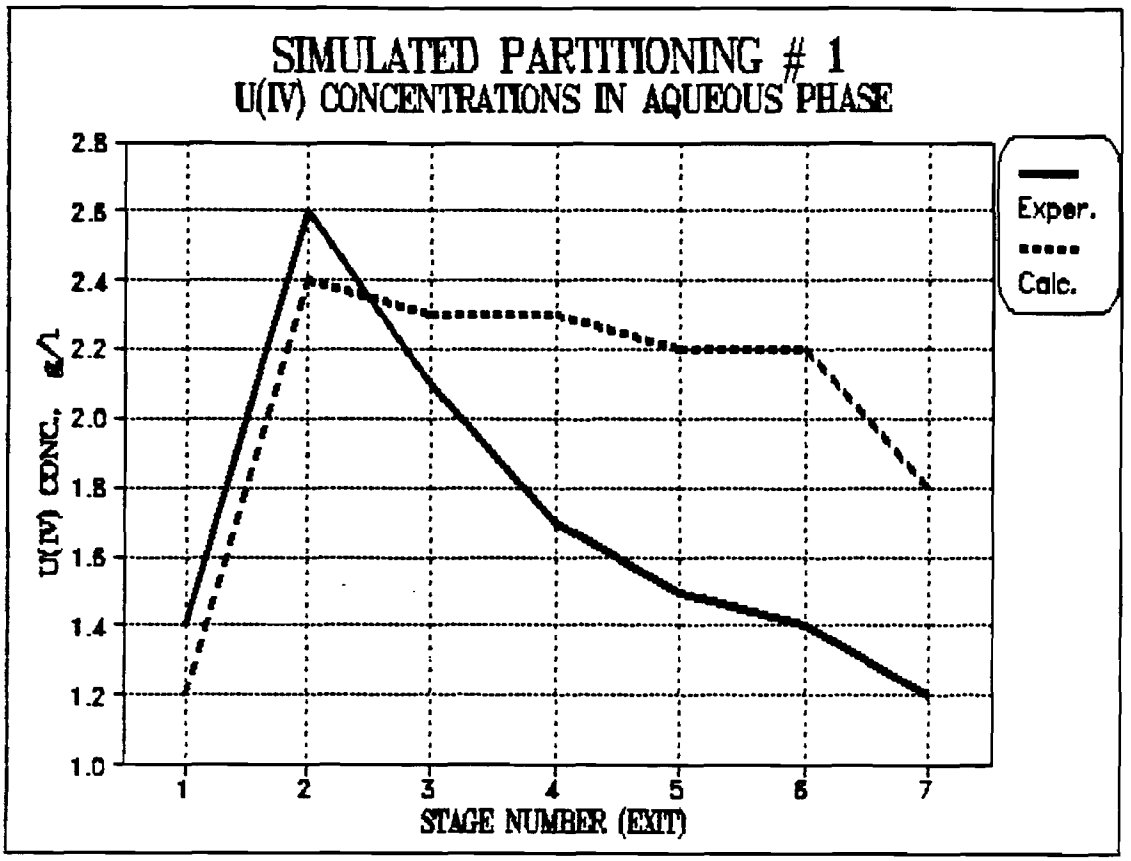


Figure 5.6 Simulated Partitioning Experiment No. 1 - U(VI) Concentration Profiles



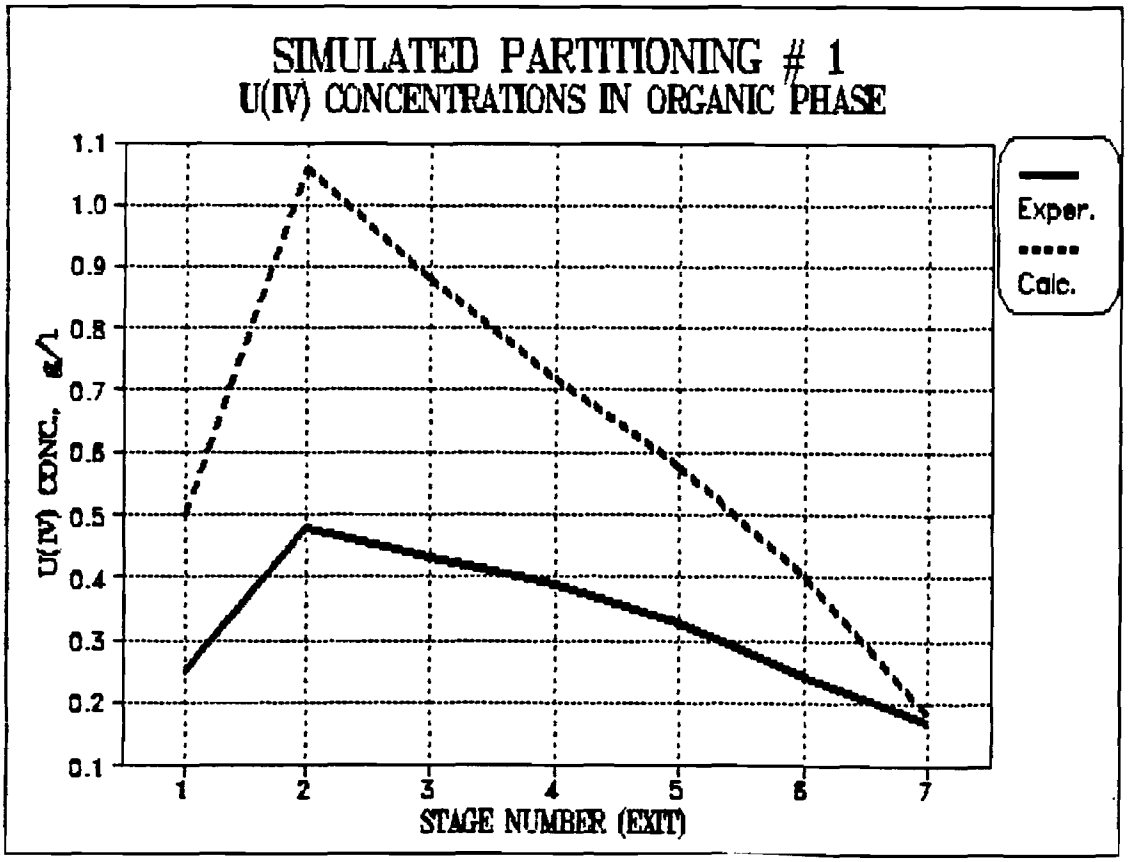


Figure 5.7 Simulated Partitioning Experiment No. 1 - U(IV) Concentration Profiles

Figure 5.7 shows that the U(IV) concentration in the organic stream was lower than expected. In addition, an increase in the aqueous U(VI) concentration, over the amount predicted by modified SEPHIS, is apparent in Figure 5.6. This increase in aqueous U(VI) concentration in the aqueous stream occurred between stages three and seven, which matches up well with the corresponding decrease in U(IV) concentration observed in Figure 5.7.

These observations led to the conclusion that the U(IV) distribution coefficients in the modified SEPHIS program were essentially correct, but that the U(IV) produced was rapidly reoxidized to U(VI). One hypothesis for the rapid U(IV) disappearance rate was that a portion of the hydrazine stabilizer in the aqueous stream was being destroyed in the electrocell, leaving the cell exit stream hydrazine deficient and the U(IV) unprotected from oxidation. A second possibility was that the U(IV) produced in the cell moved on to stage two where some fraction of it was transferred to the organic stream. The U(IV) in the organic stream was not protected by hydrazine, since it does not extract, and thus reoxidized in the presence of nitric acid and air. Some amount of this additional U(VI) in the organic may have stripped back into the aqueous, increasing the aqueous U(VI) concentration and allowing more U(IV) to be extracted into the organic phase (since the organic U(IV) concentration would have been continually decreasing from reoxidation). Both of these explanations could justify the constant U(IV) losses beyond stage three and the aqueous U(VI) increases after stage three.

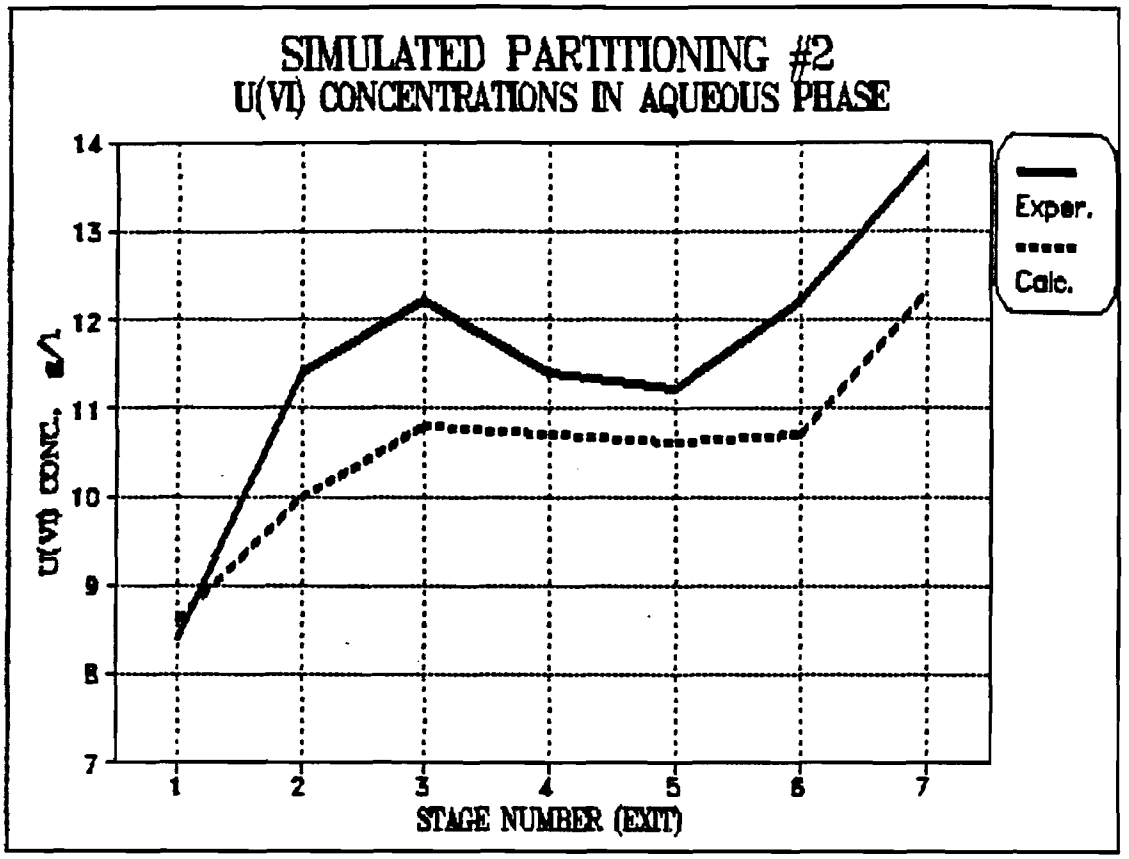
Simulated Partitioning Experiment #2:

To test the hypothesis that the consumption of hydrazine was the primary cause of the U(IV) losses in the first simulated partitioning run, a second experiment was conducted in which the initial hydrazine concentration in the aqueous was doubled (from .25 M, to .50 M). In addition, the flow rates of the organic and aqueous streams were adjusted to 17.5 and 5.7 ml/min, respectively. This resulted in an O/A ratio of roughly 3, which was more representative of an actual partitioning flow sheet.

In this experiment the initial aqueous solution was 2.55 N nitric acid and contained 0.5 M hydrazine. The organic stream had uranium (VI) and nitric acid concentrations of 82.7 g/l and .05 M, respectively. The electrocell voltage was adjusted to 2.4 volts resulting in a current of 4.9 amps. The U(VI) reduction, obtained from analyses of the aqueous stream before and after the cell, was 30%. The aqueous stream temperature at the cell exit was 27 degrees Celsius.

Comparisons of the calculated and experimental results are given in Figures 5.8 and 5.9.

The results of this experiment were consistent with the data obtained from the first simulated partitioning run. Figure 5.8 shows that the aqueous and organic U(VI) concentrations were modeled relatively well. Again, the U(VI) concentrations in the aqueous stream were slightly higher beyond the second stage than calculated. In Figure 5.9 the aqueous U(IV) concentrations



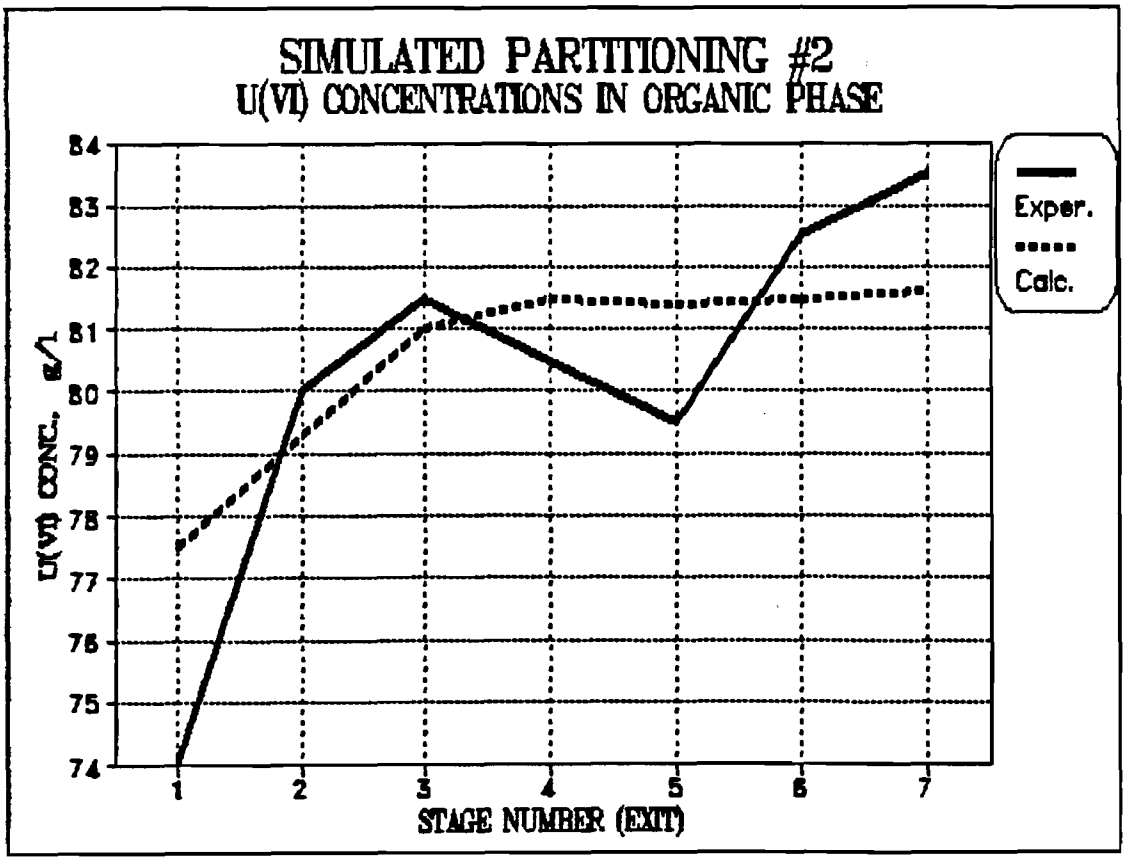
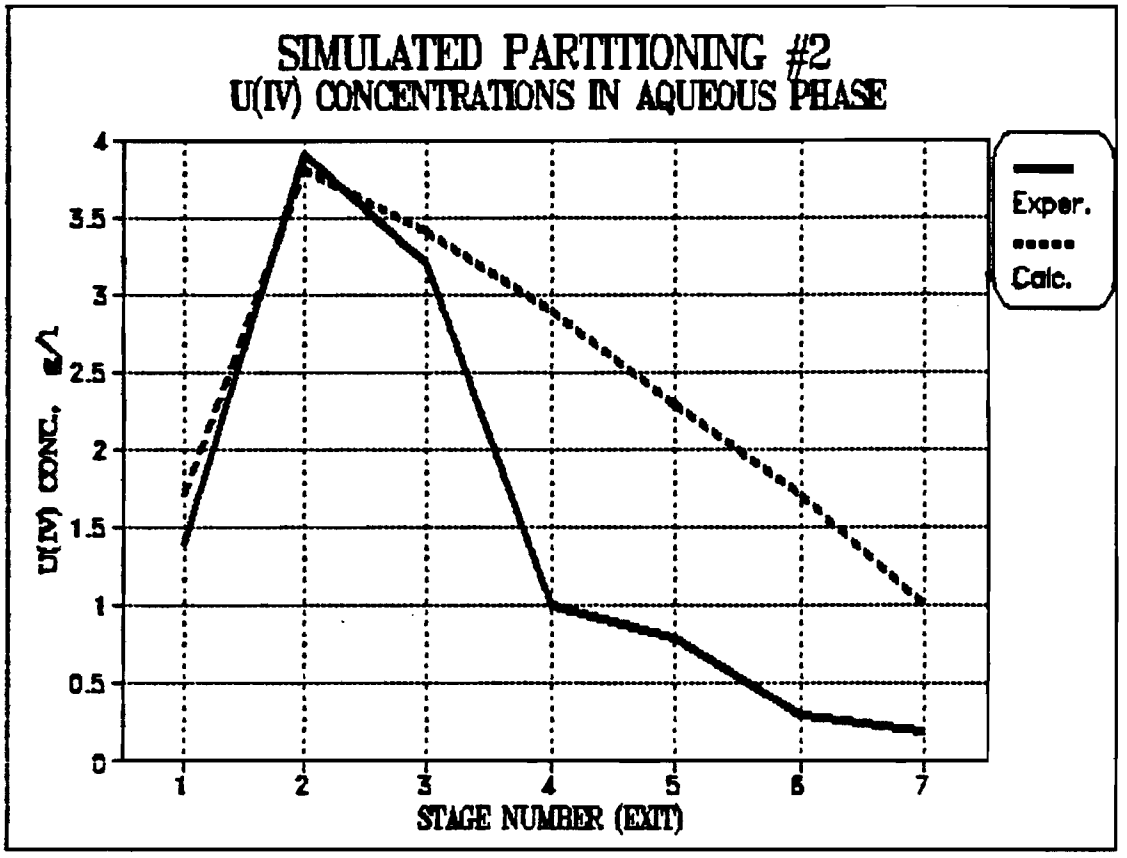


Figure 5.8 Simulated Partitioning Experiment No. 2 - U(VI) Concentration Profiles



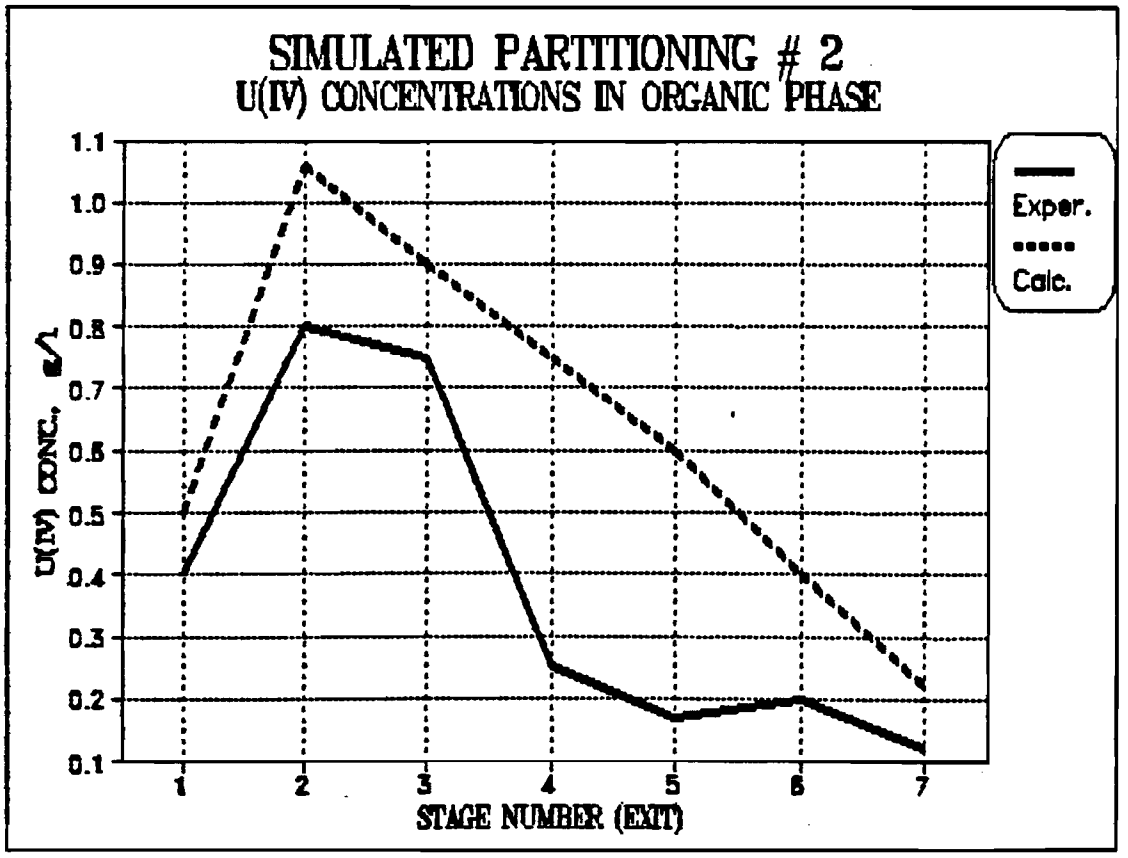


Figure 5.9 Simulated Partitioning Experiment No. 2 - U(IV) Concentration Profiles

in stages one and two are roughly equivalent to the estimates, but drop off rapidly as stage seven is approached. The results for the organic U(IV) concentration profiles were also similar to those in the previous run.

The conclusion drawn from the second simulated partitioning run was that the destruction of hydrazine was not the prime cause for the observed U(IV) losses.

Simulated Partitioning Experiment #3:

In the third experiment, an inert gas blanket was used to reduce the oxidation of U(IV) in the organic stream. Argon was chosen as the purge gas since it is inert, heavier than air, and allows for indefinite storage of U(IV) [14].

The aqueous feed was 2.55 N nitric acid and 0.53 M hydrazine. The organic solution contained 88.1 g/l U(VI) and was 0.04 N in nitric acid. The organic and aqueous flow velocities were adjusted to 17.1 and 5.7 ml/min, respectively. This gave an O/A ratio of 3. The electrocell voltage was maintained at 3.0 volts and about 5.4 amps. At that voltage an experimental reduction of about 10% was obtained.

The method used for blanketing the contactor with inert gas was to run a gas line from the argon tank to a manifold. The outlets from the manifold were then run to the individual stage gas entrance ports (normally used with nitrogen to minimize spindle corrosion). The gas was turned on slowly at the beginning of the test and argon flowed down below the spindle and exited just above the

aqueous exit port. Adjustments of the gas flow rate were necessary to avoid interfering with the contactor's normal operation. Samples were taken after 35 minutes so that any air in the system would have been flushed out.

The modified SEPHIS program was run with a cell reduction input of 10%. A comparison of the SEPHIS estimates and the experimental results are presented in Figures 5.10 and 5.11.

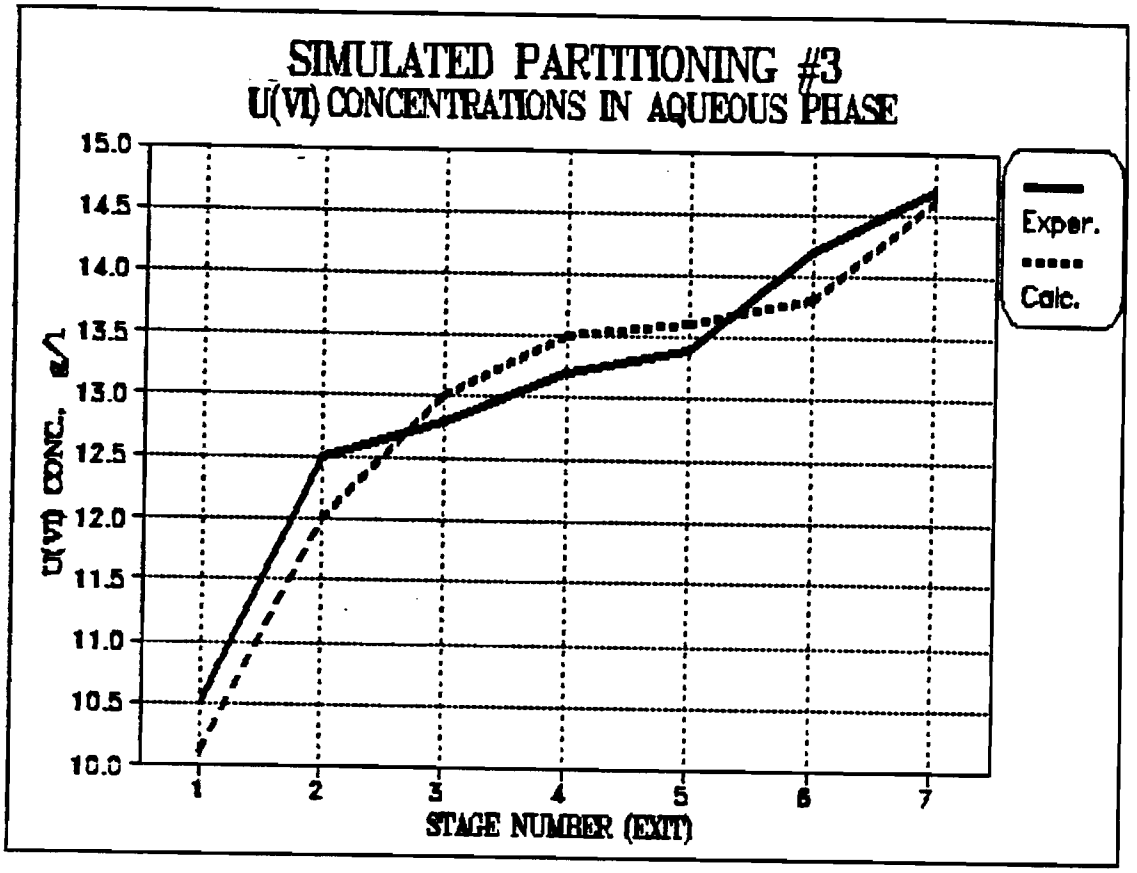
As can be seen, the problem with U(IV) losses was practically eliminated by the use of the inert gas blanketing technique. The results of the experiment showed conclusively that the modified version of SEPHIS Mod 4 could accurately model a simulated partitioning run containing U(VI) and U(IV), if extraneous U(IV) reoxidation is minimized.

5.4. Mass Balances

A mass balance was made for each of the contactor experiments. The mass balance error can be used as an overall check of the analytical techniques used for the uranium determinations.

The mass balance discrepancies were as follows:

Extraction experiment	1.6%
Stripping experiment	7.3%
Partitioning experiment #1	6.9%
Partitioning experiment #2	3.5%
Partitioning experiment #3	2.3%



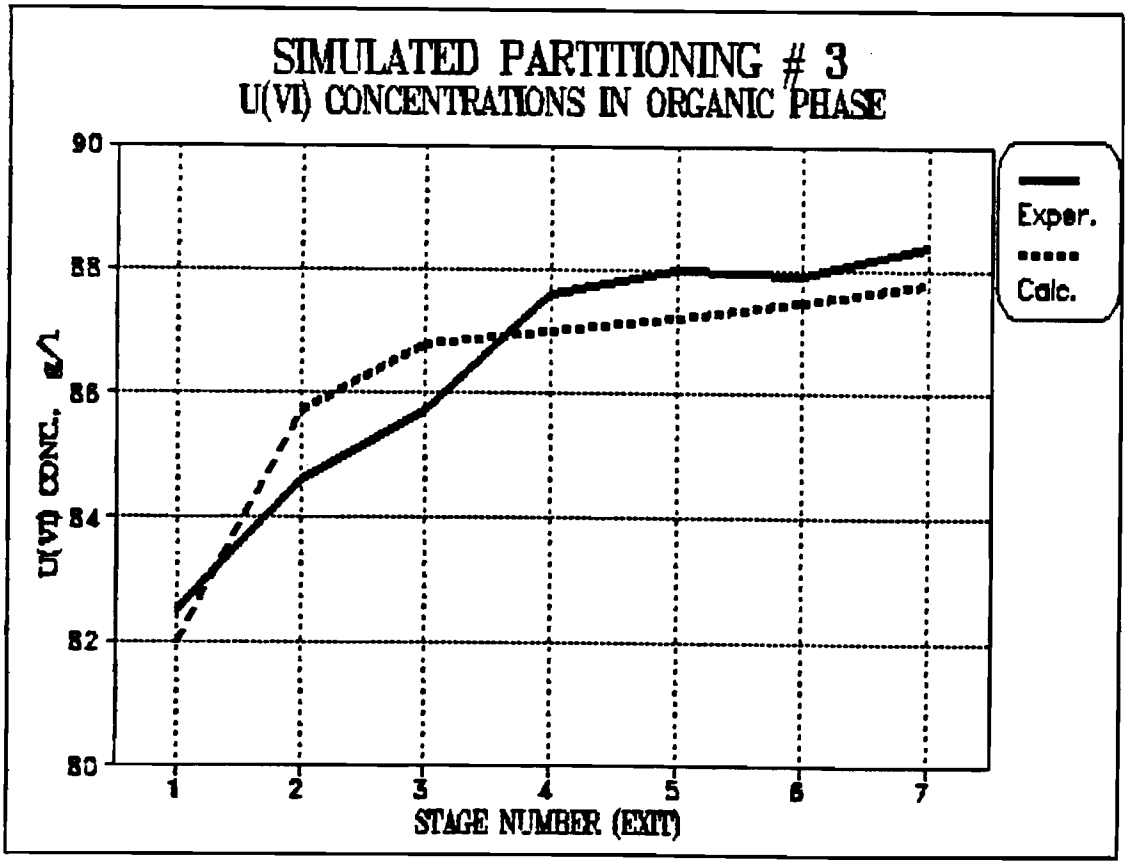
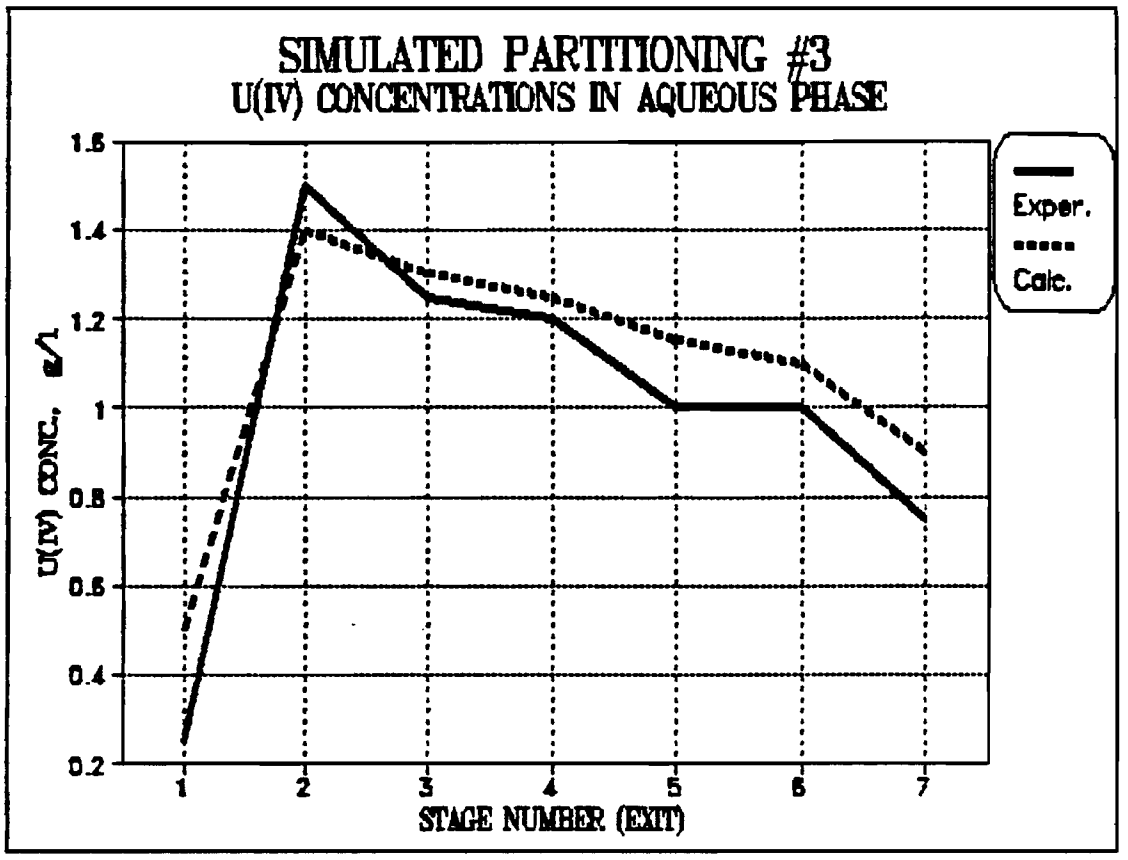


Figure 5.10 Simulated Partitioning Experiment No. 3 - U(VI) Concentration Profiles



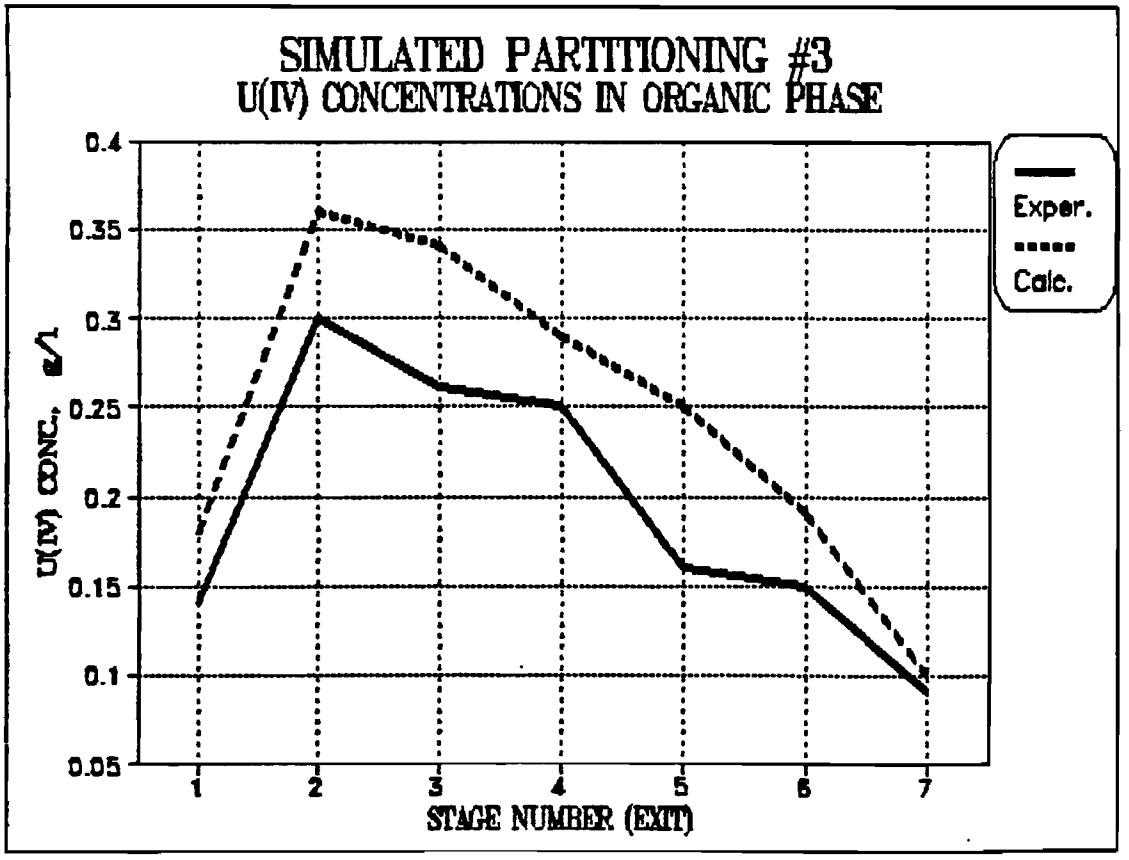


Figure 5.11 Simulated Partitioning Experiment No. 3 - U(IV) Concentration Profiles

6. SEPHIS MODIFICATION

6.1. Background

Solvent extraction is a widely used unit operation in the chemical industry. The analysis of solvent extraction systems has been based on the estimation of distribution equilibria in individual stages which could be real, as is the case in mixer settlers or centrifugal contactors, or convenient concepts for differential contactors, such as pulsed columns.

The estimation of the number of stages necessary to accomplish a certain separation and the concentration profiles throughout a cascade require stage-by-stage calculations which, because of their iterative nature, are quite lengthy and tedious. Many calculational and graphic methods have been developed for this purpose. The McCabe-Thiele graphical method was used extensively since the nineteen twenties, but it had only limited value for systems containing more than one solute. The development of solvent extraction methods in the nuclear industry in the period following World War II and the gradual availability of computers ideally suited for iterative calculations led to the development of many computer programs for multicomponent solvent extraction systems.

One of the earliest programs for the calculation of concentration profiles for multicomponent systems in mixer settlers was SOLVEX, written by J. Lowe of SRL. This program provided distributions at equilibrium as well as under transient conditions. The latter capability was particularly useful in the analysis

of upsets which might lead to criticality excursions as a consequence of the reflux of fissile nuclides in a solvent extraction contactor. A more sophisticated program, SEPHIS, written in 1972 by W. S. Groenier of ORNL and modified subsequently by G. L. Richardson and J. L. Swanson of HEDL, S. B. Watson and R. H. Rainey of ORNL, and A. D. Mitchell of ORNL has been widely used in the nuclear industry. The Mitchell version, SEPHIS-MOD4, issued in 1980 and adapted to Personal Computers by R. Jubin of ORNL in 1986, served as the basis for the development of SEPHIS MOD5GT as part of this study.

6.2. SEPHIS MOD4

The SEPHIS (Solvent Extraction Process Having Interacting Solutes) computer program by ORNL has been particularly useful for U and Pu separations with TBP solvent. The program models the steady-state and the dynamic mass transfer in multi-stage solvent extraction equipment.

The SEPHIS computer program is written in FORTRAN IV for use with IBM computing systems. It is, however, easily adapted for CDC and other machines. The computations are based on a solvent extraction contactor model consisting of a finite series of discrete stages, each containing a "perfect" mixing and settling zone. All mass transfer occurs in the mixer and plug flow is assumed. The program applies to both, extraction and stripping contactors.

The program is organized to perform the stage calculations using an iterative procedure for each stage in each time interval. The procedure used in the SEPHIS calculations makes the program applicable to several design, optimization, and evaluation problems in a solvent extraction system. Starting with any combination of concentrations of uranium, plutonium, and nitric acid, in any stage of a countercurrent contactor, and any combination of feed stream conditions and flow rates, the program calculates the step by step change in the concentrations of uranium, plutonium, and nitric acid. This type of calculation provides a means of evaluating transient, steady state, start-up, and shut-down conditions. The concentrations resulting from these calculations may be used in evaluating product losses, in determining the nuclear criticality safety of equipment, in estimating the loading of the solvent so that decontamination from fission products may be assessed, and in analyzing the effects of other procedural and operating parameters. The number of theoretical stages needed to limit the losses of U or Pu or to obtain a required separation of U and Pu may be determined by varying the number of stages until the desired results are obtained.

The flow of solutes through the apparatus being modeled is simulated by the differential equations describing the "perfect" mixers and settlers. The equations start with a transient mass balance around a mixer. Any changes in

the amount of a particular component in a stage must be equal to the difference between the amount that flows into and the amount that leaves the mixer.

Several assumptions and approximations made in SEPHIS can be grouped into the following five areas:

1. Concentrations in the contactor change relatively slowly.
2. The volumes and flowrates remain constant until changed by the user.
3. The mechanical operation of the contactor conforms with the idealities of the model.
4. Certain chemical effects or conditions are assumed to either exist or not exist.
5. Many heat effects are neglected.

The approximations necessarily lead to some differences between calculated concentrations and experimental results, but these differences can generally be localized to specific portions of a contactor.

6.3. SEPHIS MOD5GT

SEPHIS MOD5GT is a modification of SEPHIS MOD5, the PC version of SEPHIS MOD4, to permit the modeling of a solvent extraction cascade containing electrolytic reduction cells installed externally between two stages. The general structure of SEPHIS MOD5 was retained, the electroreduction being modeled by the new subroutine ELECTRO. The flow arrangements and the more important symbols used in the program are given in Figure 6.1.

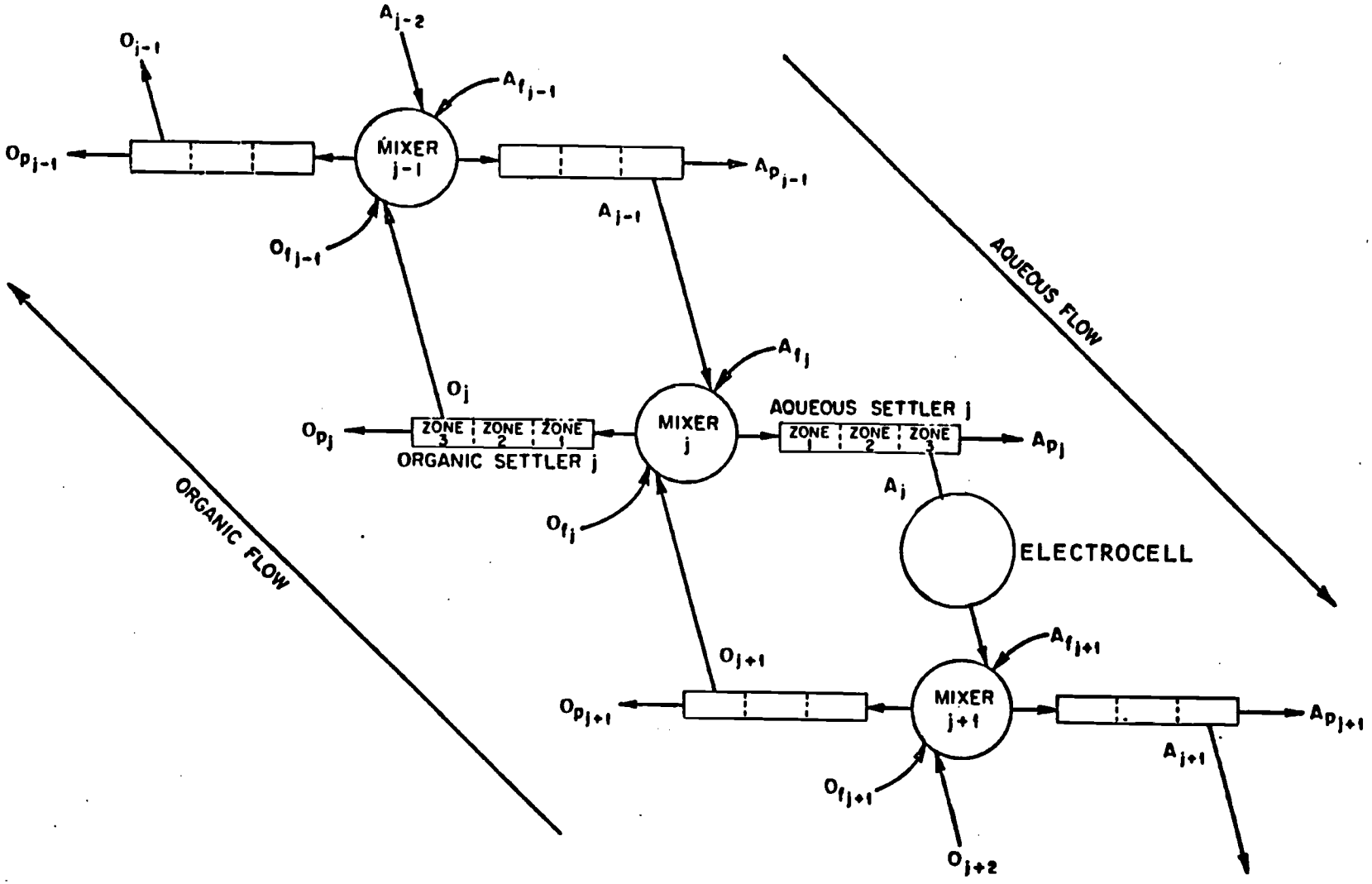


Figure 6.1 Contactor Flow Schemes and SEPHIS Symbols

The bases for ELECTRO were described in Section 5.1.2. A flow chart for ELECTRO is given in Figure 6.2. A basic flow chart for SEPHIS MOD5GT, illustrating the interaction between ELECTRO and other stages of SEPHIS MOD5, is shown in Figure 6.3.

The program was tested for three cases:

The first case had the same input as the example for the IBM mainframe version of SEPHIS MOD4 described in the user manual. The purpose of this test was to ascertain that the modifications made did not affect the performance of the program for conventional solvent extraction cascades, such as the six-stage stripping column in this example. The results shown in Appendix C are the same as those in the SEPHIS MOD4 User's Manual.

The second case models a uranium-plutonium partitioning in an eight-stage contactor with two electrocells. The parameters used for the electrocells were either empirical values from experimental data or were estimated from the dimensions of the cell and the electrodes. The program worked satisfactorily, as can be seen from the results in Appendix C.

The third example is similar to the second, except that sixteen stages were used, to determine the extent by which the losses of plutonium can be reduced in the organic exit stream. The considerable improvement is evident in the program output in Appendix C.

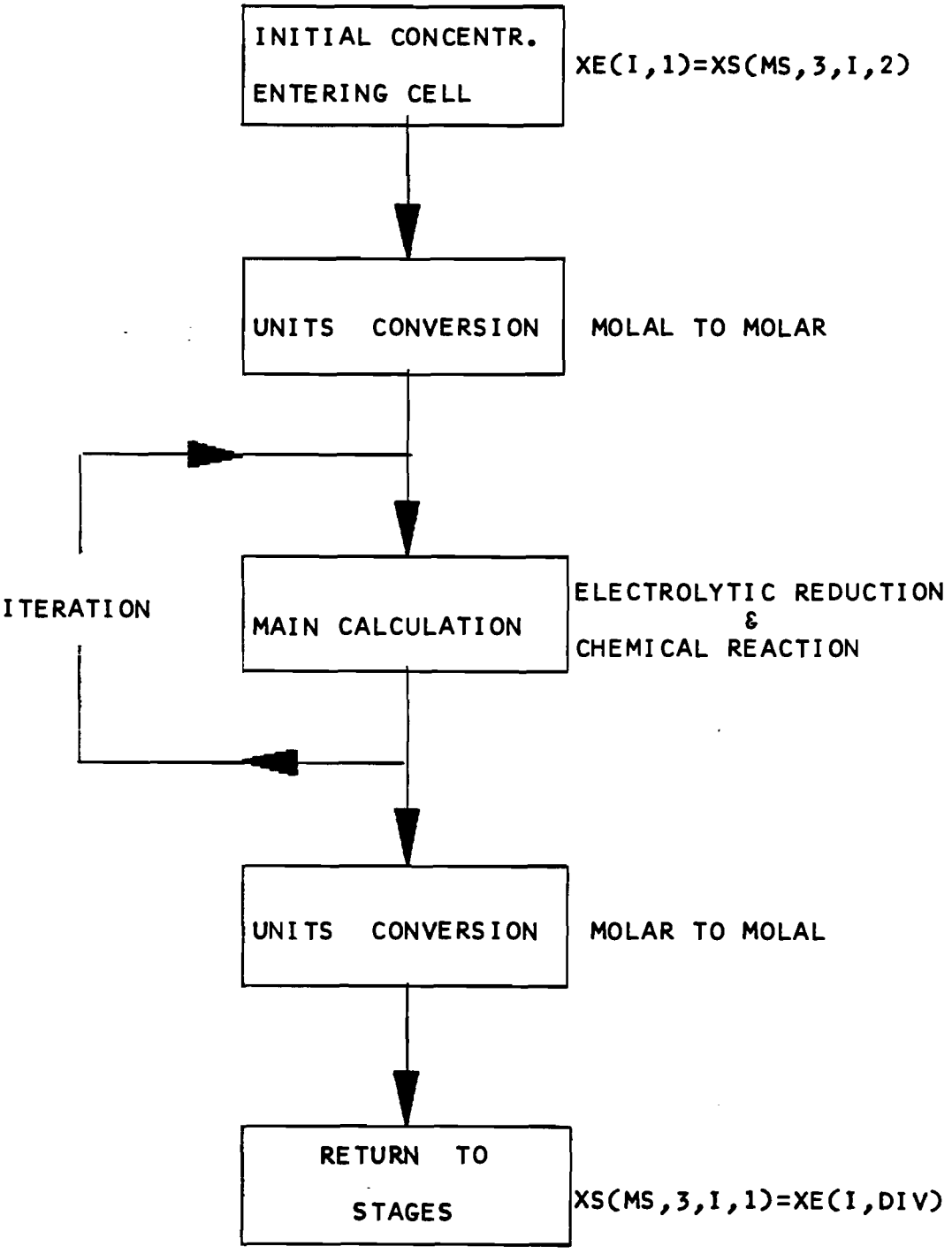


Figure 6.2 ELECTRO - Flow Chart

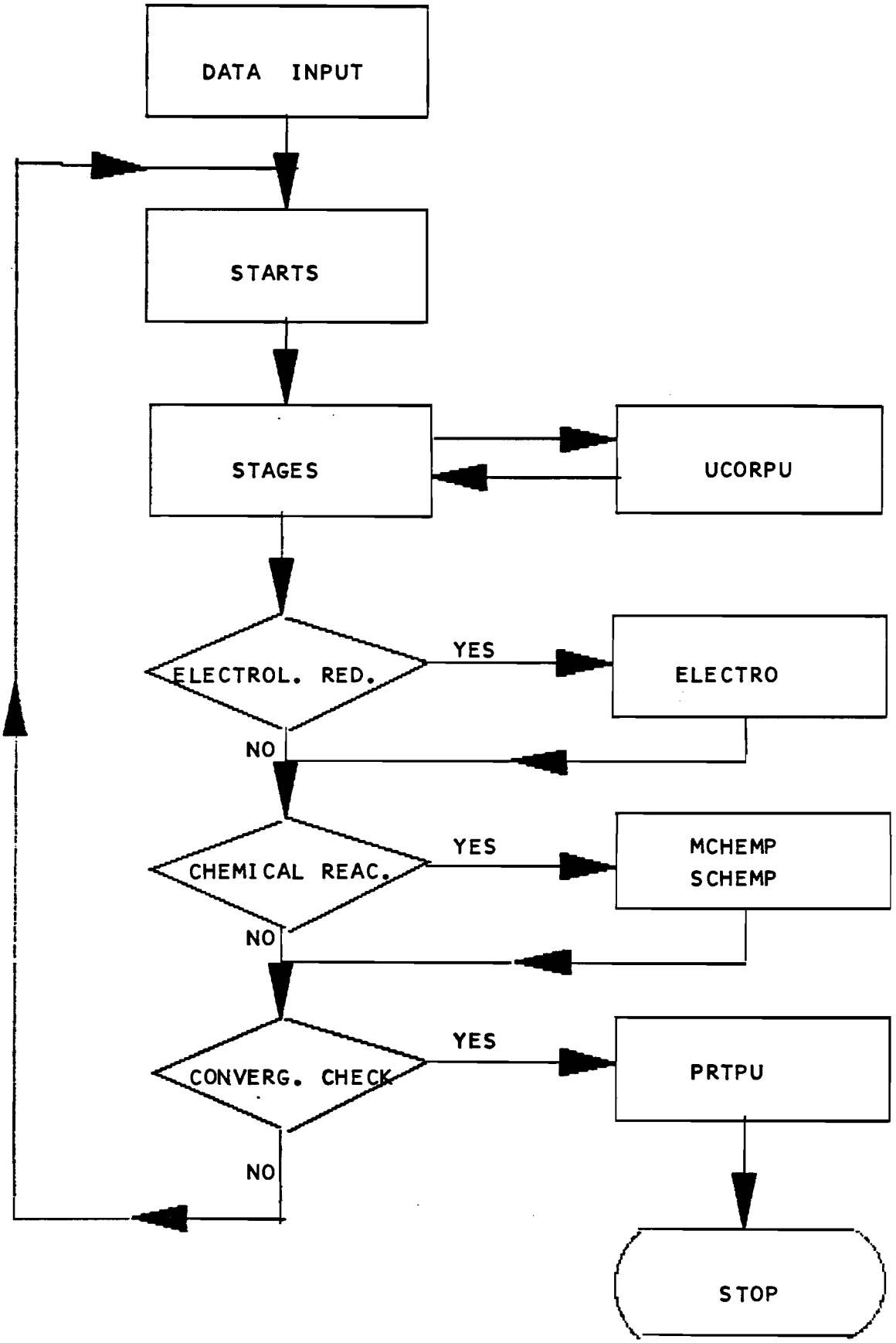


Figure 6.3 SEPHIS MOD5GT Flow Chart

Appendix C also contains the FORTRAN source file, examples of an input file and a complete description of the variables in the input file and the input format, and instructions for running SEPHIS MOD5GT. A diskette containing the complete FORTRAN source and executable files and the input and output files for the three cases described above is enclosed with this report. Because compiling of the entire program is not possible with typical PCs, the FORTRAN source file was divided into four sections which can be individually compiled; the resulting .OBJ files are then linked in the conventional manner. The four sections are also contained in the diskette.

The following running times are typical for the three examples run on three different computers:

COMPUTER TYPE	PC A	PC B	PC A
Example 1	2min 2sec	1min 2sec	0min 14sec
Example 2	18min 5sec	10min 5sec	2min 12sec
Example 3	39min 28sec	26min 10sec	5min 27sec

Computer TYPE A: IBM AT, 80286-5MHz CPU, 80287 coprocessor, 640Kb RAM.

Computer TYPE B: NCR (AT Clone), 80286-10MHz CPU, 80287 coprocessor, 640Kb RAM.

Computer TYPE C: CompuAdd (AT Clone), 80386-25MHz CPU, 80387 coprocessor, 1 Mb RAM with Cache.

7. CONCLUSIONS

1. The SEPHIS MOD 5 computer program was validated as adequately by modeling the centrifugal contactor for extraction and stripping of uranium (VI).
2. The contactor exhibited unsatisfactory hydraulic performance at O/A ratios within the organic to aqueous phase inversion region.
3. Operation of the electrocell in series with the centrifugal contactor is hydraulically feasible without compromising the contactor's performance.
4. Previous findings that uranium (VI) can be reduced electrolytically in a cell with a common electrode compartment (no diaphragm) were confirmed.
5. The "electro-reduction" cell can be used as an "electro-oxidation" cell for destroying hydrazine.
6. The ELECTRO program for modeling the electrocell was verified within the experimental ranges tested. The dependence of the U(VI) reduction rate on the initial U(VI) concentration, flow rate, and voltage was confirmed, but other parameters, not included in ELECTRO, may have to be added.
7. SEPHIS MOD5GT, consisting of a modified version of SEPHIS MOD5 and ELECTRO, modeled satisfactorily the simulated U/Pu partitioning in a PUREX flowsheet, using centrifugal contactors and in-line electrocells.

8. Hydrazine protected U(IV) from reoxidation in the aqueous phase but not in the organic phase.

9. Blanketing the centrifugal contactor with argon reduced considerably the reoxidation of U(IV).

10. Based on the results obtained with SEPHIS MOD5GT, it appears feasible to separate plutonium and uranium in an external electrocell-centrifugal contactor system.

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NOMENCLATURE

ANL	Argonne National Laboratory.
ELECTRO	Electro-reduction cell modeling program.
Ferrous Sulfamate	Chemical reductant/stabilizer used for the reduction of plutonium in the Purex process.
HAN	Hydroxylamine nitrate, a chemical reductant.
Hydrazine	A holding reductant commonly used to stabilize U(IV) solutions.
In Situ	To indicate an operation which takes place within the extraction apparatus.
LMFBR	Liquid Metal Fast Breeder Reactor.
LWR	Light Water Reactor.
NORPAR	Hydrocarbon diluent.
ORNL	Oak Ridge National Laboratory.
PUREX	Solvent extraction process employing TBP as the extractant in a hydrocarbon diluent.
SEPHIS	Program, developed at ORNL, for modeling a solvent extraction cascade.
SRL	Savannah River Laboratory.
SRP	Savannah River Plant.
TBP	Tributyl Phosphate - an extractant for uranium and plutonium.

APPENDIX A

Table A.1

Free Acid Indicator Comparison

Indicator	Uranyl Nitrate Conc. (M)	Actual Free Acid (N)	Calculated Free Acid (N)	Error (%)	Mean Error (%)	St. Dev. (%)
Congo red	.0312	.04602	.0492	+6.9%	8.85	1.95
Congo red	.0647	1.532	1.697	10.8%		
Methyl Orange	.0312	.04602	.0436	-5.3%		
Methyl Orange	.0414	3.007	3.074	+2.2%	-.4	3.47
Methyl Orange	.0647	1.532	1.561	+1.9%		
Methyl Red	.0418	1.002	1.028	+2.6%		
Methyl Red	.1063	.0891	.0879	-1.3%		
Methyl Red	.2092	1.002	.9986	-.34%		
Methyl Red	1.046	1.002	1.0084	+.64%	.18	1.33
Methyl Red	1.038	3.036	2.9996	-1.2%		
	1.083	3.0248	3.004	+.69%		

Table A.2

U(VI) Spectrophotometric Correlation Data

Phase	Acidity N	U(VI) M	Abs.(X=415nm)	*Correlation Eqn #	Correlation Coeff
A	0	.10012	.742		
A	0	.05006	.371		
A	0	.02503	.182		
A	0	.01252	.086		
				4.1	.99998
A	1	.05006	.390		
A	1	.02503	.188		
A	1	.01251	.092		
				4.2	.99992
A	3	.01780	.159		
A	3	.03380	.315		
A	3	.05254	.485		
A	3	.08552	.790		
				4.3	.99993
0	-	.00368	.027		
0	-	.01842	.141		
0	-	.03562	.252		
0	-	.04680	.382		
0	-	.09736	.758		
				4.4	.99921

*From a least squares fit

Correlation Equation

4.1 X=(Y+.0054)/7.4644 M or X=(Y+.0054)x31.885g/l
4.2 X=(Y+.009)/7.9657 M or X=(Y+.009)x29.878g/l
4.3 X=(Y+.0030)/9.28536 M or X=(Y+.0030)x25.6318g/l
4.4 X=(Y+.0074)/7.8377 M or X=(Y+.0074)x30.3699

X=U(VI) conc.
Y=Abs. reading

Table A.3

U(VI) Correlation Curve Error Analysis

Phase	Acidity	U(VI) M Actual	(X=415) Abs.	Corrl. #	U(VI) from Curve	Curve Error
A	0	.014843	.107	4.1	.0150581	-1.4%
A	0	.0238	.170	4.1	.0235	+1.03%
A	0	.03626	.263	4.1	.0359573	+.83%
A	0	.039192	.295	4.1	.0402444	-2.6%
A	0	.055358	.421	4.1	.0571245	-3.1%
A	0	.084206	.618	4.1	.0832485	+1.15%
						Avg. error ±1.69%
					Mean error ($\bar{X}$)=-1.02%	
					St. dev. (δ)=1.74%	
A	1	.015633	.119	4.2	.0160889	-2.7%
A	1	.031924	.243	4.2	.0316356	+.91%
A	1	.041416	.310	4.2	.0400467	+3.3%
A	1	.063190	.499	4.2	.0637734	-.91%
						Avg. error = ±1.96%
					Mean error ($\bar{X}$)=+.15%	
					St. dev. (δ)=2.22%	
A	3	.01264	.110	4.3	.01217	-3.7%
A	3	.02528	.231	4.3	.02520	-.32%
A	3	.04227	.382	4.3	.04147	-1.9%
A	3	.04275	.384	4.3	.04168	-2.5%
A	3	.04996	.472	4.3	.05116	+2.4%
A	3	.05056	.481	4.3	.05213	+3.1%
A	3	.07045	.651	4.3	.07044	-.01%
A	3	.09988	.942	4.3	.10178	+1.9%
						Avg. error ±1.98
					Mean error ($\bar{X}$) =-.129%	
					St. Dev. (δ)=2.3%	
0	-	.007367	.053	4.4	.007707	+4.6%
0	-	.03300	.245	4.4	.032204	-2.4%
0	-	.09209	.740	4.4	.095360	+3.6%
					Mean Error ($\bar{X}$) =+1.93%	
					St. Dev. (δ) =3.09	3.53% avg

Table A.4

U(IV) Spectrophotormetric Correlation Data

Phase	Acidity(N)	U(IV) M	Abs. Reading =648nm)	*Correl. Eng. #	Correlation Coeff.
A	**2.8 Dal Sample	.001320	.024		
A	2.8	.002955	.71		
A	2.8	.003160	.77		
A	2.8	.004433	.110		
A	2.8	.005746	.148		
				4.5	.99975
O	(10 ml Samples)	.000487	.0095		
O	"	.001462	.028		
O	"	.003139	.064		
O	"	.005672	.139		
O	"	.007731	.169		
				4.6	.9947

**.2 N hydrazine

*From a least squares fit X=U(IV) concentration
Y=Absorption reading

Correl. Equn. 4.5 X=(Y+.01215)/27.8875 M or X=(Y+.01215)x8.5343 g/l
4.6 X=(Y+.00357)/23.1108 M or X=(Y+.00357)x10.2982 g/l

Table A.5

U(IV) Correlation Curve Error Analysis

Phase	Acidity (N)	(Titration) U(IV) M Actual)	Abs.(=648nm)	Correl. #	U(IV) from Curve	Curve Error
A	2.8	.029550	.760	4.5	.027688	-6.3%
A	2.8	.003408	.108	4.5	.004433	+2.8%
A	2.8	.002735	.085	4.5	.002768	+1.1%
				Mean Error ($\bar{X}$)=-.8%		
				St. Dev. (δ) =3.95%		
				Avg. Error = 3.40		
O	-	.00656	.0122	4.6	.000682	+4.0%
O	-	.002011	.039	4.6	.001842	-8.4%
O	-	.002086	.043	4.6	.002015	-3.5%
				Mean Error ($\bar{X}$)=2.6%		
				St. Dev. (δ) =5.7%		
				Avg. Error = $\pm$5.30%		

APPENDIX B

TABLE B.1

```
      PROGRAM ELECTRO
C     PROGRAM ELECTRO CALCULATES THE CONCENTRATION PROFILES FOR
C     U(6), U(4), Pu(4), Pu(3), HNO3, AND OTHER NITRATE SALTS
C     IN AN ELECTROCELL
      REAL GAM, OC, OA, VC, FR, DT
      DIMENSION XE(6,99), DX(6,99)
      OPEN(5,FILE='F.INP')
      READ(5,10) XE(1,1), XE(2,1), XE(3,1), XE(4,1),
     *XE(5,1), XE(6,1)
   10 FORMAT(6F8.5)
      READ(5,12,END=15) BRU, BRPU, OC, OA, GAM, VC, FR, N
   12 FORMAT(5F9.6,2F8.3,/I2)
      CLOSE(5,STATUS='DELETE')
   15 DT=VC/(FR*N)
      DO 100 K=2,N
C     CALCULATE THE CONCENTRATION CHANGES DUE TO ELECTROLYTIC
C     REDUCTION OF U(6) AND Pu(4) AT THE CATHODE AND THE
C     CHEMICAL REACTION BETWEEN U(4) AND Pu(4).
      DX(2,K)=-XE(2,K-1)*(1-EXP(-BRU*OC*DT))+GAM*
     *XE(5,K-1)*XE(3,K-1)/XE(1,K-1)**2*DT
      XE(2,K)=XE(2,K-1)+DX(2,K)
      DX(5,K)=-DX(2,K)
      XE(5,K)=XE(5,K-1)+DX(5,K)
      DX(3,K)=-XE(3,K-1)*(1-EXP(-BRPU*OC*DT))-2*GAM*XE(5,K-1)*
     *XE(3,K-1)/XE(1,K-1)**2*DT
      XE(3,K)=XE(3,K-1)+DX(3,K)
      IF(XE(3,K).LT.0) XE(3,K)=0.0
      DX(4,K)=-DX(3,K)
      XE(4,K)=XE(4,K-1)+DX(4,K)
      DX(1,K)=2*DX(2,K)
      XE(1,K)=XE(1,K-1)+DX(1,K)
  100 CONTINUE
      OPEN(6,FILE='F.OUT')
      WRITE(6,180)
  180 FORMAT(1X,' CONCENTRATIONS AT ENTRANCES TO SUB-CELLS'//)
      WRITE(6,200)
  200 FORMAT(1X,'SUB-CELL #  U(6),M     U(4),M     Pu(4),M',
     *'     Pu(3),M    HNO3,M       MNO3,M'/)
      DO 300 K=1,N
      WRITE(6,210) K, XE(2,K), XE(5,K), XE(3,K), XE(4,K),
     *XE(1,K), XE(6,K)
  210 FORMAT(4X,I3,6F12.5)
  300 CONTINUE
      CLOSE(6,STATUS='KEEP')
      END
```

TABLE B.2

Sample ELECTRO Input

```
 2.55000 0.04412 0.00000 0.00000 0.00210 0.53000
 0.00795  0.000    1.95     0.0      0.0      47.6     5.700
60
```

Line 1: HNO_3 conc., U(VI) conc., Pu(IV) conc., Pu(III) conc., U(IV) conc., and nitrate concentration.

Line 2: β (U reduction), β (Pu reduction), Ω (cathode). Ω (anode), δ (reaction rate constant), cell volume, flow rate.

Line 3: Number of subdivisions.

Sample ELECTRO Output

CONCENTRATIONS AT ENTRANCES TO SUB-CELLS

SUB-CELL #	U(6),M	U(4),M	Pu(4),M	Pu(3),M	HNO3,M	MNO3,M
1	0.04412	0.00210	0.00000	0.00000	2.55000	0.53000
60	0.03885	0.00737	0.00000	0.00000	2.53945	0.00000

TABLE B.3

Effect of Subcell Number

CONCENTRATIONS AT ENTRANCES TO SUB-CELLS

SUB-CELL #	U(6),M	U(4),M	Pu(4),M	Pu(3),M	HNO3,M	MNO3,M
1	0.04412	0.00210	0.00000	0.00000	2.55000	0.53000
99	0.03881	0.00741	0.00000	0.00000	2.53939	0.00000

SUB-CELL #	U(6),M	U(4),M	Pu(4),M	Pu(3),M	HNO3,M	MNO3,M
1	0.04412	0.00210	0.00000	0.00000	2.55000	0.53000
40	0.03889	0.00733	0.00000	0.00000	2.53954	0.00000

SUB-CELL #	U(6),M	U(4),M	Pu(4),M	Pu(3),M	HNO3,M	MNO3,M
1	0.04412	0.00210	0.00000	0.00000	2.55000	0.53000
20	0.03901	0.00721	0.00000	0.00000	2.53979	0.00000

SUB-CELL #	U(6),M	U(4),M	Pu(4),M	Pu(3),M	HNO3,M	MNO3,M
1	0.04412	0.00210	0.00000	0.00000	2.55000	0.53000
10	0.03927	0.00695	0.00000	0.00000	2.54030	0.00000

SUB-CELL #	U(6),M	U(4),M	Pu(4),M	Pu(3),M	HNO3,M	MNO3,M
1	0.04412	0.00210	0.00000	0.00000	2.55000	0.53000
5	0.03978	0.00644	0.00000	0.00000	2.54132	0.00000

SUB-CELL #	U(6),M	U(4),M	Pu(4),M	Pu(3),M	HNO3,M	MNO3,M
1	0.04412	0.00210	0.00000	0.00000	2.55000	0.53000
3	0.04047	0.00575	0.00000	0.00000	2.54270	0.00000

TABLE B.4

SEPHIS Output - Extraction Experiment

```
CALCULATIONS FOR A SOLVENT EXTRACTION PROCESS HAVING INTERACTING SOLUTES


PUREX PROCESS
EXPERIMENT-BY D.Pschirer
THIS IS AN EXPERIMENTAL TESTING OF A SEPHIS MODIFICATION
CASE Ext.1 - STEADY STATE RUN,
-DTHETA =    1.000 MINUTES PER TIME INCREMENT
DPRINT = 1000.000 MINUTES BETWEEN PRINTING OF PROFILES
IFAST = 1  THE FAST INTEGRATION TECHNIQUE WILL BE USED
THIS TIME PERIOD WILL END WHEN TIME = TSTOP = 1000.000 MINUTES, OR A TOLERANCE OF TOL =   .0100 % PER MINUTE IS REACHED
NUMBER OF STAGES =  7
NEWIN = 1 NEW INPUT FLOWS WILL BE GIVEN
NEWOUT = 1  NEW OUTPUT FLOWS WILL BE GIVEN
IVOLM = 3 MIXER VOLUMES DETERMINED BY PHASE FLOW
IVOLS = 3 SETTLER VOLUMES GIVEN BY PHASE FLOW
IPRO = 0  A NEW INITIAL PROFILE WILL NOT BE READ
IPNCH = 0   NO PUNCHED CARD OUTPUT
NSTR = 0   NO UNUSUAL ROUTING PATTERN
TEMPI =  2.500E+01  INITIAL & DEFAULT TEMPERATURE
IRXN = 4  EXPERMENT AS SUGGESTED BY DR. SCHNEIDER
STAGE EFFICIENCY OF 1.00
REDUCTION EFFICIENCY OF  .00
-FEED & PRODUCT      STAGE  NITRIC ACID    U (VI)       PU (IV)      PU (III)      U (IV)     NITRATE ION    FLOW RAT      TEMP
STREAM DATA           NO.     (MOL/L)      (G/L)        (G/L)        (G/L)        (G/L)       (MOL/L)      (L/MIN)        (C)
          AQUEOUS    1    7.000E-01    9.860E+01    .000E+00     .000E+00     .000E+00     .000E+00    1.220E+01      25.0
          AQUEOUS    2    1.000E-01    9.860E+01    .000E+00     .000E+00     .000E+00     .000E+00    1.220E+01      25.0
       30.0 % TBP    7    4.000E-02    3.400E+00    .000E+00     .000E+00     .000E+00     .000E+00    1.210E+01      25.0
          AQUEOUS    1    PRODUCT STREAM REMOVED (FLOW RATE IN MOLAL UNITS)                             1.220E+01


- TIME =  292.00  MINUTES
    AQUEOUS PHASE
        STAGE ^ NITRIC ACID^  U (VI)   ^  PU (IV)   ^  PU (III)  ^  U (IV)   ^ NITRATE ION^  DENSITY   ^ MIXER FLOW ^TEMPERATURE
         NO.  ^   (MOL/L)  ^   (G/L)   ^   (G/L)    ^   (G/L)    ^   (G/L)    ^   (MOL/L)  ^   (G/ML)   ^   (L/MIN)  ^ (CENTIGRAE)
              ^            ^           ^            ^            ^            ^            ^            ^            ^
            1 ^ 1.008E-01 ^ 9.205E+01 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ 1.129E+00 ^ 1.218E+01 ^ 2.500E+01
            1 ^ PRODUCT STREAM                                                                   ^ 1.218E+01 ^
            2 ^ 1.015E-01 ^ 8.012E+01 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ 1.112E+00 ^ 1.213E+01 ^ 2.500E+01
            3 ^ 1.031E-01 ^ 6.328E+01 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ 1.088E+00 ^ 1.207E+01 ^ 2.500E+01
            4 ^ 1.043E-01 ^ 4.592E+01 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ 1.064E+00 ^ 1.200E+01 ^ 2.500E+01
            5 ^ 7.049E-01 ^ 3.215E+01 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ 1.045E+00 ^ 1.195E+01 ^ 2.500E+01
            6 ^ 1.076E-01 ^ 2.230E+01 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ 1.031E+00 ^ 1.192E+01 ^ 2.500E+01
            7 ^ 1.369E-01 ^ 1.355E+01 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ 1.020E+00 ^ 1.190E+01 ^ 2.500E+01
    ORGANIC PHASE
        STAGE ^ NITRIC ACID^  U (VI)   ^  PU (IV)   ^  PU (III)  ^  U (IV)   ^ NITRATE ION^  DENSITY   ^ MIXER FLOW ^ INVENTORY
         NO.  ^   (MOL/L)  ^   (G/L)   ^   (G/L)    ^   (G/L)    ^   (G/L)    ^   (MOL/L)  ^   (G/ML)   ^   (L/MIN)  ^  CHANGE %)
              ^            ^           ^            ^            ^            ^            ^            ^            ^
            1 ^ 5.541E-03 ^ 8.697E+01 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ 9.369E-01 ^ 1.244E+01 ^ 1.604E-03
            2 ^ 6.108E-03 ^ 8.054E+01 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ 9.278E-01 ^ 1.241E+01 ^ 4.333E-03
            3 ^ 7.069E-03 ^ 6.884E+01 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ 9.114E-01 ^ 1.236E+01 ^ 7.150E-03
            4 ^ 8.136E-03 ^ 5.231E+01 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ 8.885E-01 ^ 1.229E+01 ^ 9.516E-03
            5 ^ 8.801E-03 ^ 3.526E+01 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ 8.650E-01 ^ 1.222E+01 ^ 9.927E-03
            6 ^ 9.057E-03 ^ 2.172E+01 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ 8.466E-01 ^ 1.216E+01 ^ 8.348E-03
            7 ^ 1.143E-02 ^ 1.201E+01 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ .000E+00 ^ 8.335E-01 ^ 1.212E+01 ^ 5.270E-03
```

TABLE B.5

SEPHIS Output - Stripping Experiment

STAGE EFFICIENCY OF 1.00
REDUCTION EFFICIENCY OF .00

FEED & PRODUCT STREAM DATA	STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	FLOW RAT (L/MIN)	TEMP (C)
AQUEOUS	1	1.000E-02	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	1.570E+01	25.0
AQUEOUS	2	1.000E-02	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	1.570E+01	25.0
30.0 % TBP	7	8.000E-02	8.480E+01	.000E+00	.000E+00	.000E+00	.000E+00	1.300E+01	25.0
AQUEOUS	1	PRODUCT STREAM REMOVED (FLOW RATE IN MOLAL UNITS)						1.570E+01	

AQUEOUS PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	TEMPERATURE (CENTIGRAE)
1	1.024E-02	4.284E+00	.000E+00	.000E+00	.000E+00	.000E+00	1.003E+00	1.572E+01	2.500E+01
1	PRODUCT STREAM							1.572E+01	
2	1.010E-02	1.490E+01	.000E+00	.000E+00	.000E+00	.000E+00	1.018E+00	1.577E+01	2.500E+01
3	1.010E-02	2.642E+01	.000E+00	.000E+00	.000E+00	.000E+00	1.033E+00	1.583E+01	2.500E+01
4	1.006E-02	3.676E+01	.000E+00	.000E+00	.000E+00	.000E+00	1.048E+00	1.588E+01	2.500E+01
5	1.018E-02	4.661E+01	.000E+00	.000E+00	.000E+00	.000E+00	1.061E+00	1.593E+01	2.500E+01
6	1.334E-02	5.735E+01	.000E+00	.000E+00	.000E+00	.000E+00	1.076E+00	1.598E+01	2.500E+01
7	7.406E-02	6.835E+01	.000E+00	.000E+00	.000E+00	.000E+00	1.094E+00	1.607E+01	2.500E+01

ORGANIC PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	INVENTORY CHANGE %
1	2.510E-04	3.256E-01	.000E+00	.000E+00	.000E+00	.000E+00	8.176E-01	1.259E+01	9.758E-03
2	5.621E-04	5.668E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.247E-01	1.261E+01	6.022E-03
3	7.386E-04	1.889E+01	.000E+00	.000E+00	.000E+00	.000E+00	8.425E-01	1.267E+01	3.758E-03
4	7.778E-04	3.318E+01	.000E+00	.000E+00	.000E+00	.000E+00	8.619E-01	1.273E+01	2.028E-03
5	7.720E-04	4.598E+01	.000E+00	.000E+00	.000E+00	.000E+00	8.795E-01	1.279E+01	1.162E-03
6	9.562E-04	5.815E+01	.000E+00	.000E+00	.000E+00	.000E+00	8.963E-01	1.284E+01	6.628E-04
7	4.901E-03	7.137E+01	.000E+00	.000E+00	.000E+00	.000E+00	9.149E-01	1.290E+01	3.443E-04

TABLE B.6

SEPHIS Output - Simulated Partitioning Experiment #1

STAGE EFFICIENCY OF 1.00
REDUCTION EFFICIENCY OF .35

FEED & PRODUCT STREAM DATA	STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	FLOW RAT (L/MIN)	TEMP (C)
AQUEOUS	1	2.800E+00	.000E+00	.000E+00	.000E+00	.000E+00	2.500E-01	1.570E+01	25.0
AQUEOUS	2	2.800E+00	.000E+00	.000E+00	.000E+00	.000E+00	2.500E-01	1.570E+01	25.0
30.0 % TBP	7	2.000E-01	0.540E+01	.000E+00	.000E+00	.000E+00	.000E+00	1.300E+01	25.0
AQUEOUS	1	PRODUCT STREAM REMOVED (FLOW RATE IN MOLAL UNITS)						1.570E+01	

- TIME = 118.08 MINUTES

AQUEOUS PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	TEMPERATURE (CENTIGRAE)
1	2.766E+00	5.789E+00	.000E+00	.000E+00	4.668E-01	2.498E-01	1.154E+00	1.571E+01	2.500E+01
1	PRODUCT STREAM							1.571E+01	
2	2.771E+00	7.910E+00	.000E+00	.000E+00	2.382E+00	2.494E-01	1.158E+00	1.574E+01	2.500E+01
3	2.759E+00	1.002E+01	.000E+00	.000E+00	2.330E+00	2.493E-01	1.161E+00	1.574E+01	2.500E+01
4	2.751E+00	1.137E+01	.000E+00	.000E+00	2.295E+00	2.493E-01	1.162E+00	1.575E+01	2.500E+01
5	2.746E+00	1.235E+01	.000E+00	.000E+00	2.263E+00	2.492E-01	1.164E+00	1.575E+01	2.500E+01
6	2.741E+00	1.314E+01	.000E+00	.000E+00	2.191E+00	2.492E-01	1.165E+00	1.575E+01	2.500E+01
7	2.724E+00	1.386E+01	.000E+00	.000E+00	1.874E+00	2.493E-01	1.165E+00	1.574E+01	2.500E+01

ORGANIC PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	INVENTORY CHANGE %)
1	3.212E-01	6.627E+01	.000E+00	.000E+00	1.779E-01	.000E+00	9.181E-01	1.298E+01	1.789E-03
2	2.834E-01	7.320E+01	.000E+00	.000E+00	7.424E-01	.000E+00	9.267E-01	1.299E+01	3.333E-03
3	2.566E-01	7.818E+01	.000E+00	.000E+00	6.128E-01	.000E+00	9.329E-01	1.300E+01	3.186E-03
4	2.428E-01	8.072E+01	.000E+00	.000E+00	5.500E-01	.000E+00	9.361E-01	1.300E+01	4.378E-03
5	2.340E-01	8.234E+01	.000E+00	.000E+00	5.093E-01	.000E+00	9.381E-01	1.301E+01	6.155E-03
6	2.276E-01	8.353E+01	.000E+00	.000E+00	4.703E-01	.000E+00	9.396E-01	1.301E+01	8.277E-03
7	2.216E-01	8.448E+01	.000E+00	.000E+00	3.844E-01	.000E+00	9.407E-01	1.301E+01	9.335E-03

TABLE B.7

SEPHIS Output - Simulated Partitioning Experiment #3

STAGE EFFICIENCY OF 1.00
REDUCTION EFFICIENCY OF .10

FEED & PRODUCT STREAM DATA	STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	FLOW RAT (L/MIN)	TEMP (C)
AQUEOUS	1	2.550E+00	.000E+00	.000E+00	.000E+00	.000E+00	5.300E-01	5.700E+00	25.0
AQUEOUS	2	2.550E+00	.000E+00	.000E+00	.000E+00	.000E+00	5.300E-01	5.700E+00	27.0
30.0 % TBP	7	4.000E-02	8.810E+01	.000E+00	.000E+00	.000E+00	.000E+00	1.710E+01	25.0
AQUEOUS	1	PRODUCT STREAM REMOVED (FLOW RATE IN MOLAL UNITS)						5.700E+00	

- TIME = 166.00 MINUTES

AQUEOUS PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	TEMPERATURE (CENTIGRAE)
1	2.494E+00	1.087E+01	.000E+00	.000E+00	6.158E-01	5.290E-01	1.209E+00	5.711E+00	2.585E+01
1	PRODUCT STREAM							5.711E+00	
2	2.521E+00	1.330E+01	.000E+00	.000E+00	1.577E+00	5.279E-01	1.214E+00	5.723E+00	2.663E+01
3	2.517E+00	1.408E+01	.000E+00	.000E+00	1.480E+00	5.279E-01	1.215E+00	5.723E+00	2.629E+01
4	2.514E+00	1.419E+01	.000E+00	.000E+00	1.352E+00	5.279E-01	1.215E+00	5.722E+00	2.598E+01
5	2.498E+00	1.416E+01	.000E+00	.000E+00	1.169E+00	5.282E-01	1.214E+00	5.719E+00	2.570E+01
6	2.418E+00	1.428E+01	.000E+00	.000E+00	9.168E-01	5.297E-01	1.211E+00	5.703E+00	2.545E+01
7	2.050E+00	1.565E+01	.000E+00	.000E+00	6.082E-01	5.360E-01	1.199E+00	5.636E+00	2.521E+01

ORGANIC PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	INVENTORY CHANGE %)
1	2.302E-01	8.213E+01	.000E+00	.000E+00	1.611E-01	.000E+00	9.376E-01	1.719E+01	3.699E-03
2	2.132E-01	8.568E+01	.000E+00	.000E+00	3.654E-01	.000E+00	9.421E-01	1.721E+01	3.153E-03
3	2.070E-01	8.684E+01	.000E+00	.000E+00	3.249E-01	.000E+00	9.435E-01	1.721E+01	4.821E-03
4	2.056E-01	8.710E+01	.000E+00	.000E+00	2.925E-01	.000E+00	9.439E-01	1.721E+01	6.727E-03
5	2.045E-01	8.714E+01	.000E+00	.000E+00	2.502E-01	.000E+00	9.439E-01	1.721E+01	8.370E-03
6	1.989E-01	8.715E+01	.000E+00	.000E+00	1.893E-01	.000E+00	9.438E-01	1.720E+01	9.336E-03
7	1.702E-01	8.727E+01	.000E+00	.000E+00	1.048E-01	.000E+00	9.429E-01	1.718E+01	8.829E-03

APPENDIX C

INSTRUCTIONS FOR RUNNING SEP5GT

1. Load SEP5GT.EXE onto a hard disk or blank formatted diskette.

2. Create an input file ‹name›.INP, following the format given in this Apendix, and copy this file to the directory containing SEPT5GT.EXE.

3. Designate ‹name›.OUT as the output file.

4. Enter: SEP5GT ‹name›.INP ‹name›.OUT

5. Running of the program is indicated by a time output on the screen. Note that these are "contactor running times" and not the computer program running times.

6. The results will be found in file ‹name›.OUT in the directory containing SEPT5GT.EXE.

```
  SEPHIS MOD VGT
  FORTRAN PROGRAM LISTING

NOFLOATCALLS
STORAGE:2
DEBUG

  THE ABOVE THREE LINES ARE MICROSOFT COMPILER OPTIONS

  PROGRAM SEPHIS
     GEORGIA TECH SEP. 1988       MODIFIED BY  A. SCHNEIDER & K. K. LI
     ORNL, JAN. 1986              PC VERSION BY R. T. JUBIN
     ORNL, APRIL 10, 1980         REVISIONS BY A.D.MITCHELL
     ORNL REVISION OF THE RAINEY-WATSON MODIFIED VERSION (ORNL-TM-5123)
     WATSON & RAINEY MODIFIED THE RICHARDSON-NANCE VERSION (HEDL-TME 75-31)
     ORIGINAL PROGRAM BY W.S.GROENIER (ORNL 4746)

     TRANSIENT STAGE CALCULATIONS FOR PUREX SOLVENT EXTRACTION PROCESS.
     CALCULATIONS DONE FOR 6 COMPONENTS
          5 TRANSFERRING COMPONENTS        COMPONENT 1 - NITRIC ACID
                                           COMPONENT 2 - URANIUM (VI)
                                           COMPONENT 3 - PLUTONIUM (IV)
                                           COMPONENT 4 - PLUTONIUM (III)
                                           COMPONENT 5 - URANIUM (IV)
          1 NON-TRANSFERRING COMPONENT     COMPONENT 6 - NITRATE ION

     100 STAGES ALLOWED, FOR 100 STAGES SET NTOST = 00


     DESCRIPTION OF MOST MAJOR VARIABLES
         AVOL   VOLUME OF AQUEOUS PHASE IN MIXER J
         OVOL   VOLUME OF ORGANIC PHASE IN MIXER J
         ASVOL  VOLUME OF AQUEOUS PHASE IN SETTLER J
         OSVOL  VOLUME OF ORGANIC PHASE IN SETTLER J
         TPROF  MIXER TEMPERATURE
         ATS    AQUEOUS TEMPERATURE IN THE SETTLER
         OTS    ORGANIC TEMPERATURE IN THE SETTLER
         XFD (I,J)   CONCENTRATION OF COMPONENT I IN AQUEOUS FEED TO STAGE J
         YFD (I,J)   CONCENTRATION OF COMPONENT I IN ORGANIC FEED TO STAGE J
         AFDRT   FLOW RATE (LITERS/MIN) OF AQUEOUS FEED TO MIXER J
         OFDRT   FLOW RATE (LITERS/MIN) OF ORGANIC FEED TO MIXER J
         AFDTEM  TEMPERATURE OF AQUEOUS FEED TO MIXER J
         OFDTEM  TEMPERATURE OF ORGANIC FEED TO MIXER J
         X (I,J,K)  AQUEOUS CONCENTRATION OF COMPONENT I IN MIXER J DURING
                    A TIME INTERVAL K   K=1  FOR PREVIOUS TIME
                                        K=2  FOR PRESENT TIME
         XS (J,L,I,K) AQUEOUS CONC OF COMPONENT I IN SETTLER J, ZONE L, DURING
                    A TIME INTERVAL K.  EACH SETTLER IS SPLIT INTO THREE
                    ZONES IN ORDER TO SMOOTH OUT THE CHANGES IN CONCENTRATION
                    OF ITS MIXER.  THE ZONES CAN BE THOUGHT OF AS WELL
                    MIXED TANKS FLOWING INTO THE NEXT ZONE OR MIXER.
         A      AQUEOUS INTERSTAGE FLOW RATE
         O      ORGANIC INTERSTAGE FLOW RATE
         AT     AQUEOUS INTRASTAGE FLOW RATE
         OT     ORGANIC INTRASTAGE FLOW RATE
         ALVRT  FLOW RATE OF AQUEOUS STREAM LEAVING MIXER-SETTLER BANK
         OLVRT  FLOW RATE OF ORGANIC STREAM LEAVING MIXER-SETTLER BANK
         AOUT, OOUT   INTERSTAGE FLOW RATES IN REAL VOLUME UNITS,
                      USED ONLY IN PRINT-OUT
         NSOLU  IS THE SUBSCRIPT FOR THE HIGHEST NUMBERED COMPONENT
                IN THE SYSTEM.  IT IS USED IN THE SAME MANNER AS ISOL.
         ISOL   INDICATES IF A SOLUTE IS (OR WAS) PRESENT IN THE
                CONTACTOR.  IT IS USED TO BYPASS USELESS CALCULATIONS
```

```
C               OF ZERO CONCENTRATION.
C          SPH   SPECIFIC HEAT OF ORGANIC PHASE  (AQUEOUS ASSUMED TO BE 1)
C          NDIREC DETERMINES THE ORDER OF THE STAGEWISE CALCULATIONS.
C          NDIREC = 1 FOR CALCULATIONS TO START AT THE FIRST STAGE
C                 =-1 FOR CALCULATIONS TO START WITH THE LAST STAGE
C          CODUM       LARGEST CHANGE IN SOLUTE INVENTORY OF A COMPONENT
C                      IN THE MIXER
C
C   TITLE IS THE PROBLEM TITLE.
C   NTOST = TOTAL NUMBER OF STAGES, MUST NOT EXCEED 100
C    CTBP = VOLUME FRACTION OF DRY TBP
C   TEMPI = AN INITIAL OR DEFAULT TEMPERATURE
C    NSTR = 0 NO UNUSUAL ROUTING PATTERN DESIRED
C         = 1 ROUTING PATTERN OTHER THAN NORMAL WILL BE USED
C             NSTR, ISTR, JSTR  VARIABLES SPECIFYING THAT THE ORGANIC PHASE
C                               OF STAGE ISTR BE ROUTED TO STAGE JSTR
C    IRXN = INDICATOR OF THE REACTION USED FOR REDUCTION
C         = 0  NO REACTIONS WILL BE CONSIDERED
C         = 1  INSTANTANEOUS REDUCTION OF PLUTONIUM
C         = 2  REDUCTION OF PLUTONIUM BY URANIUM (IV)
C         = 3  REDUCTION OF PLUTONIUM BY HYDROXYLAMINE
C         = 4  ELECTROCELL WITHOUT CHEMICAL REACTION
C         = 5  ELECTROCELL PLUS REDUCTION BY HYDROXYLAMINE
C         = 6  ELECTROCELL PLUS REDUCTION BY U(IV)
C         = 7  ELECTROCELL PLUS INSTANTANEOUS REDUCTION
C  DTHETA = TIME INCREMENT (MINUTES)
C  DPRINT = ELAPSED TIME BETWEEN THE PRINTING OF PROFILES
C   TSTOP = TIME WHEN THE TIME PERIOD IS TO END
C     TOL = THE TOLERANCE TO BE USED TO TEST FOR CONVERGENCE
C   NEWIN = 0 IF THE PRESENT INPUT STREAMS ARE TO BE CONTINUED
C         = 1 IF NEW INPUT STREAMS ARE TO BE SPECIFIED
C  NEWOUT = 0 IF THE PRESENT EXITING STREAMS ARE TO BE CONTINUED
C         = 1 IF NEW EXITING STREAMS ARE TO BE SPECIFIED
C   IVOLM = 1 IF AQUEOUS AND ORGANIC MIXER VOLUMES ARE TO BE GIVEN
C         = 2 IF TOTAL MIXER VOLUME IS GIVEN WITH PHASE VOLUMES DETERMINED
C             BY PHASE FLOW IN THE MIXER
C         = 3 IF ALL MIXER VOLUMES GIVEN BY PHASE FLOW IN THE MIXER*UNIT TIME
C   IVOLS = 1 IF AQUEOUS AND ORGANIC SETTLER VOLUMES ARE TO BE GIVEN
C         = 2 IF TOTAL SETTLER VOLUME IS GIVEN WITH PHASE VOLUMES DETERMINED
C             BY PHASE FLOW IN THE MIXER
C         = 3 IF ALL SETTLER VOLUMES GIVEN BY PHASE FLOW IN THE MIXER*UNIT TIME
C    IPRO = 0 FOR A ZERO INITIAL CONCENTRATION PROFILE
C           1 FOR A NON-ZERO INITIAL PROFILE
C   IFAST = 0  IF THE RUNGE KUTTA INTEGRATION IS TO BE USED
C         = 1  IF THE TRAPEZOIDAL INTEGRATION METHOD IS TO BE USED
C         = 2 IF THE NEW INTEGRATION METHOD IS TO BE USED
C   IPNCH = 0 IF NO PUNCHED CARD OUTPUT DESIRED
C         = 1 FOR FINAL PROFILE OUTPUT ON PUNCHED CARDS
C
CCCCCC
      COMMON/CONTRL/ NTOST,NSOLU,CTBP,NEWIN,NEWOUT,SPH,IPNCH,IFAST,IRXN,
     .  DTHETA,IVOLM,IVOLS,IPRO,TEMPI,ISOL(6),NSTR,ISTR,JSTR,CODUM(100)
      COMMON/CONCS/ X(6,100,2),Y(6,100,2),XS(100,3,6,2),YS(100,3,6,2)
      COMMON/FLOWS/ A(100),O(100),AT(100),OT(100)
      COMMON/STREAM/ XFD(6,100),YFD(6,100),AFDRT(100),OFDRT(100),
     .   AFDTEM(100),OFDTEM(100),ALVRT(100),OLVRT(100)
      COMMON/TEMPS/ TPROF(100,2),ATS(100,3,2),OTS(100,3,2)
      COMMON/VOLS/ AVOL(100),OVOL(100),ASVOL(100),OSVOL(100)
      COMMON/ELECT/ XP(6,100),XE(6,99),DX(6,99),FR,DIV,DT,
     .              PS,QS,MS,VC,BRU,BRPU,OC,OA,GAM
      INTEGER PS,QS,DIV
      REAL*8 TITLE(10,10)
      OPEN(21,FILE=' ')
```

```
      OPEN(25,FILE='FOR25.DAT',STATUS='NEW')
      OPEN(7,FILE='CON')
      NDIREC=1

NEXT CASE IS NEW

 10   CONTINUE
      READ (21,3000)NTTL,NTOST,CTBP,TEMPI,NSTR,ISTR,JSTR,IRXN
      IF(IRXN.LE.0) IRXN=0
IF IRXN.LT.4 , THE CHARACTERISTICS OF ELECTROCELL WILL BE BYPASSED
      IF(IRXN.LT.4) GO TO 11
      READ (21,3001)PS,QS,DIV,VC,BRU,BRPU,OC,OA,GAM
 11   DO 12 J=1,NTTL
         READ (21,1000)(TITLE(J,N),N=1,10)
 12   CONTINUE
      IF(NTOST.LE.0) NTOST =100
      NSOLU=6
      SPH=0.321+0.078*CTBP
      DO 14 I=1,NSOLU
         ISOL(I)=0
 14   CONTINUE

ZEROING OF ARRAYS

      DO 16 J=1,NTOST
         CODUM(J)=200.
         AVOL(J)=0.0
         OVOL(J)=0.0
         ASVOL(J)=0.0
         OSVOL(J)=0.0
         AFDRT(J) = 0.0
         OFDRT(J) = 0.0
         AFDTEM(J)=TEMPI
         OFDTEM(J)=TEMPI
         A(J) = 0.0
         O(J) = 0.0
         AT(J)=0.0
         OT(J)=0.0
         ALVRT(J) = 0.0
         OLVRT(J) = 0.0
         DO 15 I=1,NSOLU
            X(I,J,1)=0.0
            X(I,J,2)=0.0
            Y(I,J,1)=0.0
            Y(I,J,2)=0.0
            XFD(I,J) = 0.0
            YFD(I,J) = 0.0
 15      CONTINUE
 16   CONTINUE
      DO 20 L=1,2
         DO 19 J=1,NTOST
            TPROF(J,L)=TEMPI
            DO 18 K=1,3
               ATS(J,K,L)=TEMPI
               OTS(J,K,L)=TEMPI
               DO 17 M=1,NSOLU
                  XS(J,K,M,L)=0.0
                  YS(J,K,M,L)=0.0
 17            CONTINUE
 18         CONTINUE
 19      CONTINUE
 20   CONTINUE
CCC
```

```
C THIS IS THE START OF A NEW TIME PERIOD
C    ANY ONE, OR GROUP OF THESE VARIABLES MAY BE CHANGED FOR THE NEW TIME
C    PERIOD.
C    IF DTHETA=0.0 THE PRESENT CASE IS ENDED AND A NEW ONE BEGUN DEPENDING
C    ON DPRINT.  IF DPRINT=1.0, A NEW CASE IS STARTED AND A NEW TITLE READ
C    IN.  IF DPRINT=0.0, THE PROGRAM STOPS.
C
CCCC
   21 READ(21,3005)DTHETA,DPRINT,TSTOP,TOL,NEWIN,NEWOUT,IVOLM,IVOLS,
     .  IPRO,IFAST,IPNCH
      IF(DTHETA.EQ.0.) GO TO 400
      IF(NEWIN+NEWOUT+IVOLM+IVOLS+IPRO.NE.0) GO TO 23
      PRTIME=PRTIME+DPRINT
      WRITE (6,3201)DTHETA
      WRITE (6,3202)DPRINT
      IF(IFAST.EQ.1) WRITE (6,5000)
      IF(IFAST.NE.1.AND.IFAST.NE.2) WRITE (6,5001)
      WRITE (6,3203)TSTOP,TOL
      GO TO 140
   23 WRITE (6,1002)
      ICOUNT = 0
      WRITE (6,9000)
      DO 25 J=1,NTTL
         WRITE (6,1003)(TITLE(J,I),I=1,10)
   25 CONTINUE
      WRITE (6,3201)DTHETA
      WRITE (6,3202)DPRINT
      IF(IFAST.EQ.1) WRITE (6,5000)
        IF(IFAST.EQ.2) WRITE (6,5002)
      IF(IFAST.NE.1.AND.IFAST.NE.2) WRITE (6,5001)
      WRITE (6,3203)TSTOP,TOL
      WRITE (6,2000)NTOST


C  SCAN INPUT SWITCHES FOR ERRORS

      IF(NEWIN.EQ.1) WRITE (6,3101)
      IF(NEWIN.EQ.0) WRITE (6,3102)
      IF(NEWIN.NE.1.AND.NEWIN.NE.0) WRITE (6,3109)NEWIN
      IF(NEWOUT.EQ.1) WRITE (6,3103)
      IF(NEWOUT.EQ.0) WRITE (6,3104)
      IF(NEWOUT.NE.1.AND.NEWOUT.NE.0) WRITE (6,3110)NEWOUT
      IF(IVOLM.EQ.0) WRITE (6,3105)
      IF(IVOLM.EQ.1) WRITE (6,2038)
      IF(IVOLM.EQ.2) WRITE (6,2039)
      IF(IVOLM.EQ.3) WRITE (6,2040)
      IF(IVOLM.LT.0.OR.IVOLM.GT.3) WRITE (6,2041)IVOLM
      IF(IVOLS.EQ.0) WRITE (6,3106)
      IF(IVOLS.EQ.1) WRITE (6,2042)
      IF(IVOLS.EQ.2) WRITE (6,2043)
      IF(IVOLS.EQ.3) WRITE (6,2044)
      IF(IVOLS.LT.0.OR.IVOLS.GT.3) WRITE (6,2045)IVOLS
      IF(IPRO.EQ.0) WRITE (6,3107)
      IF(IPRO.EQ.1) WRITE (6,3108)
      IF(IPRO.NE.0.AND.IPRO.NE.1) WRITE (6,2003)IPRO
      IF(IPNCH.EQ.0) WRITE (6,2026)
      IF(IPNCH.EQ.1) WRITE (6,3111)
      IF(IPNCH.NE.0.AND.IPNCH.NE.1) WRITE (6,2028)IPNCH
      IF(NSTR.EQ.0) WRITE (6,2034)
      IF(NSTR.EQ.1) WRITE (6,2035)
      IF(NSTR.NE.0.AND.NSTR.NE.1) WRITE (6,2036)NSTR
```

```
      IF(NSTR.EQ.1) WRITE (6,4000)ISTR,JSTR
      WRITE (6,7000)TEMPI
      IF(IRXN.EQ.0) WRITE (6,8000)
      IF(IRXN.EQ.1) WRITE (6,8001)
      IF(IRXN.EQ.2) WRITE (6,8002)
      IF(IRXN.EQ.3) WRITE (6,8003)
      IF(IRXN.EQ.4) WRITE (6,8004)
      IF(IRXN.EQ.5) WRITE (6,8005)
      IF(IRXN.EQ.6) WRITE (6,8006)
      IF(IRXN.EQ.7) WRITE (6,8007)

    PRINT THE CHARACTERISTICS OF THE ELECTROCELLS

      IF(IRXN.LT.4) GO TO 31
      WRITE (6,8010) PS,PS+1
      WRITE (6,8010) QS,QS+1
      WRITE (6,8011) VC
      WRITE (6,8012) BRU
      WRITE (6,8013) BRPU
      WRITE (6,8014) OC
      WRITE (6,8015) OA
      WRITE (6,8016) GAM

     ZERO INITIAL CONCENTRATIONS IN ELECTROCELL

      DO 30 I=1,6
         XP(I,PS)=0.0
         XP(I,QS)=0.0
         DO 28 J=1,DIV
            XE(I,J)=0.0
 28      CONTINUE
 30   CONTINUE

   SUBROUTINE STARTS DOES ALL THE INPUT REQUIRED
   BEFORE THE ITERATIONS START

   31 CALL STARTS


      WRITE (6,3205)
      TOTIME=0.0
      PRTIME=0.0
      DO 50 J=1,NTOST
      DO 50 I=1,NSOLU
      X(I,J,2)=X(I,J,1)
      Y(I,J,2)=Y(I,J,1)
   50 CONTINUE
      GO TO 200
  140 CONTINUE

   SUBROUTINE STAGES PERFORMS STAGE CALCULATIONS FOR EACH TIME INTERVAL

      CALL STAGES (NDIREC)


      NDIREC=-NDIREC

   MERELY INCREMENTING TIME FOR TEMPS & CONCS

      TOTIME=TOTIME+DTHETA
      NSTOP=0
      DO 150 J=1,NTOST
      IF(CODUM(J).GT.TOL) NSTOP=1
```

```
      TPROF(J,1)=TPROF(J,2)
      DO 148 KZ=1,3
      ATS(J,KZ,1)=ATS(J,KZ,2)
148   OTS(J,KZ,1)=OTS(J,KZ,2)
      DO 151 I=1,NSOLU
      IF(ISOL(I).EQ.0) GO TO 149
      X(I,J,1)=X(I,J,2)
      Y(I,J,1)=Y(I,J,2)
      DO 152 KZ=1,3
      XS(J,KZ,I,1)=XS(J,KZ,I,2)
      YS(J,KZ,I,1)=YS(J,KZ,I,2)
152   CONTINUE
149   CONTINUE
151   CONTINUE
150   CONTINUE


 CHECK FOR CONVERGENCE

      IF(NSTOP.EQ.0) TSTOP=TOTIME
200   CONTINUE
      WRITE(7,999) TOTIME
999   FORMAT('+ELAPSED TIME IS ',F7.2)
      IF(TOTIME.LT.PRTIME.AND.TOTIME.LT.TSTOP) GO TO 140
      PRTIME=PRTIME+DPRINT


 PRINTING CONCENTRATIONS FOR THE TIME INTERVAL
 SUBROUTINE PRTOUT DOES ALL THE CONVERSIONS AND CALCULATIONS WHICH
 ARE REQUIRED BEFORE THE PROFILE CAN BE PRINTED OR PUNCHED.  BECAUSE
 OF THE HEADINGS, THE PRINTING IS DONE IN PRTOUT WHILE THE PUNCHING
 CAN BE DONE IN THE MAIN PROGRAM.

      WRITE (6,1100)TOTIME
      CALL PRTPU
      IF(ICOUNT.EQ.1) WRITE (6,1203)
      IF(ICOUNT.EQ.1) GO TO 201
      IF(ICOUNT.EQ.0) ICOUNT = 1
      GOTO 202
201   ICOUNT = 0
202   CONTINUE


250   IF(TOTIME.LT.TSTOP) GO TO 140


  FLOPPY DISK OUTPUT

      IF(IPNCH.EQ.0) GO TO 300
      ONE=1.0

      DO 275 J=1,NTOST
      WRITE (25,1202) (X(I,J,2),I=1,6),TPROF(J,2)
      WRITE (25,1202) (Y(I,J,2),I=1,6),ONE
      WRITE (6,1202) (X(I,J,2),I=1,6),TPROF(J,2)
      WRITE (6,1202) (Y(I,J,2),I=1,6),ONE
275   CONTINUE

300   CONTINUE
      PRTIME = PRTIME-DPRINT
      GO TO 21
400   CONTINUE
      IF(DPRINT.EQ.1.) GO TO 10
```

```
  420 CONTINUE
      CLOSE(21)
      CLOSE(25)
      STOP


 1000 FORMAT(10A8)
 1002 FORMAT('  CALCULATIONS FOR A SOLVENT EXTRACTION PROCESS',
     .  ' HAVING INTERACTING SOLUTES'//)
 1003 FORMAT(' ',10A8)
 1202 FORMAT(1P8E10.3)
 1203 FORMAT('  ')
 1100 FORMAT ('-  TIME = ',0PF7.2,'  MINUTES')
 2000 FORMAT(' NUMBER OF STAGES = ',I3)
 2003 FORMAT(' IPRO = ',I3,3X,'INVALID VALUE')
 2026 FORMAT(' IPNCH = 0',3X,'NO PUNCHED CARD OUTPUT')
 2028 FORMAT(' IPNCH = ',I3,3X,'INVALID VALUE')
 2034 FORMAT(' NSTR = 0',3X,'NO UNUSUAL ROUTING PATTERN')
 2035 FORMAT(' NSTR = 1',3X,'ROUTING PATTERN OTHER THAN NORMAL')
 2036 FORMAT(' NSTR = ',I3,3X,'INVALID VALUE')
 2038 FORMAT(' IVOLM = 1 AQUEOUS AND ORGANIC MIXER VOLUMES GIVEN')
 2039 FORMAT(' IVOLM = 2 TOTAL MIXER VOLUME GIVEN')
 2040 FORMAT(' IVOLM = 3 MIXER VOLUMES DETERMINED BY PHASE FLOW')
 2041 FORMAT(' IVOLM =',I2,' INVALID VALUE')
 2042 FORMAT(' IVOLS = 1 AQUEOUS AND ORGANIC SETTLER VOLUMES GIVEN')
 2043 FORMAT(' IVOLS = 2 TOTAL SETTLER VOLUME GIVEN')
 2044 FORMAT(' IVOLS = 3 SETTLER VOLUMES GIVEN BY PHASE FLOW')
 2045 FORMAT(' IVOLS =',I2,' INVALID VALUE')
 3000 FORMAT(2I2,2F8.0,4I2)
 3001 FORMAT(I2,I2,I3,F8.3,5F9.6)
 3005 FORMAT(4E8.0,8I2)
 3101 FORMAT(' NEWIN = 1 NEW INPUT FLOWS WILL BE GIVEN')
 3102 FORMAT(' NEWIN = 0 INPUT FLOWS WILL BE UNCHANGED')
 3103 FORMAT(' NEWOUT = 1  NEW OUTPUT FLOWS WILL BE GIVEN')
 3104 FORMAT(' NEWOUT = 0  OUTPUT STREAMS WILL BE UNCHANGED')
 3105 FORMAT(' IVOLM = 0  MIXER VOLUMES WILL BE UNCHANGED')
 3106 FORMAT(' IVOLS = 0  SETTLER VOLUMES WILL BE UNCHANGED')
 3107 FORMAT(' IPRO = 0  A NEW INITIAL PROFILE WILL NOT BE READ')
 3108 FORMAT(' IPRO = 1  A NEW INITIAL PROFILE WILL BE READ')
 3109 FORMAT(' NEWIN = ',I2,' INVALID VALUE')
 3110 FORMAT(' NEWOUT = ',I2,' INVALID VALUE')
 3111 FORMAT(' IPNCH = 1  PROFILE DATA WILL BE STORED AT END OR TIME')
 3201 FORMAT('-DTHETA = ',0PF8.3,' MINUTES PER TIME INCREMENT')
 3202 FORMAT(' DPRINT =',0PF9.3,' MINUTES BETWEEN PRINTING OF PROFILES')
 3203 FORMAT(' THIS TIME PERIOD WILL END WHEN TIME = TSTOP = ',0PF8.3,
     . ' MINUTES, OR A TOLERANCE OF TOL =',0PF8.4,' % PER MINUTE IS REAC
     .HED')
 3205 FORMAT('  TRANSIENT BEHAVIOR RESULTS   MIXER CONCENTRATION GIVEN')
 4000 FORMAT(' ORGANIC STREAM EXITING AT STAGE',I4,' FEEDS STAGE',I4)
 5000 FORMAT(' IFAST = 1  THE FAST INTEGRATION TECHNIQUE WILL BE USED')
 5001 FORMAT(' IFAST = 0  THE RUNGE-KUTTA INTEGRATION WILL BE USED')
 5002 FORMAT(' IFAST = 2  THE NEW INTEGRATION METHOD')
 7000 FORMAT(' TEMPI = ',1PE10.3,'  INITIAL & DEFAULT TEMPERATURE')
 8000 FORMAT(' IRXN = 0  NO REACTIONS WILL BE CONSIDERED')
 8001 FORMAT(' IRXN = 1  INSTANTANEOUS REDUCTION OF PLUTONIUM')
 8002 FORMAT(' IRXN = 2  REDUCTION OF PLUTONIUM BY URANIUM (IV)')
 8003 FORMAT(' IRXN = 3  REDUCTION OF PLUTONIUM BY HYDROXYLAMINE ')
 8004 FORMAT(' IRXN = 4  ELECTROCELL WITHOUT CHEMICAL REACTION')
 8005 FORMAT(' IRXN = 5  ELECTROCELL PLUS REDUCTION BY HYDROXYLAMINE')
 8006 FORMAT(' IRXN = 6  ELECTROCELL PLUS REDUCTION BY U(IV)')
 8007 FORMAT(' IRXN = 7  ELECTROCELL PLUS INSTANTANEOUS REDUCTION')
 8010 FORMAT(' ELECTROCELL BETWEEN STAGE ',I2,' AND STAGE ',I2)
 8011 FORMAT(' VC   = ',F7.4,' LITER -- VOLUME OF ELECTROCELL')
```

```
8012 FORMAT(' BRU  = ',F7.4,'       -- ELECTRICAL FACTOR FOR U(VI)')
8013 FORMAT(' BRPU = ',F7.4,'       -- ELECTRICAL FACTOR FOR PU(IV)')
8014 FORMAT(' OC   = ',F7.4,'       -- GEOMETRIC FACTOR OF CATHODE')
8015 FORMAT(' OA   = ',F7.4,'       -- GEOMETRIC FACTOR OF ANODE')
8016 FORMAT(' GAM  = ',F7.4,'       -- REACTION RATE CONSTANT FOR U(IV)
    .+ PU(IV) REACTION'/)
9000 FORMAT(' PUREX PROCESS')
     END
     SUBROUTINE CONVPU
```

SUBROUTINE CONVRT PRINTS THE FEED STREAM INFORMATION, AND CONVERTS THE CONCENTRATIONS FROM THEIR ORIGINAL FORM TO THE MOLAL UNITS WHICH ARE USED IN THE CALCULATIONS.

```
     COMMON/CONTRL/ NTOST,NSOLU,CTBP,NEWIN,NEWOUT,SPH,IPNCH,IFAST,IRXN,
    .  DTHETA,IVOLM,IVOLS,IPRO,TEMPI,ISOL(6),NSTR,ISTR,JSTR,CODUM(100)
     COMMON/CONCS/ X(6,100,2),Y(6,100,2),XS(100,3,6,2),YS(100,3,6,2)
     COMMON/MOLALC/ AQ(6),OR(6),TEMP,CONVA,CONVO,TCONC
     COMMON/STREAM/ XFD(6,100),YFD(6,100),AFDRT(100),OFDRT(100),
    .   AFDTEM(100),OFDTEM(100),ALVRT(100),OLVRT(100)
     COMMON/TEMPS/ TPROF(100,2),ATS(100,3,2),OTS(100,3,2)
```

IF NEW INPUT STREAMS HAVE BEEN GIVEN (NEWIN = 1), THIS SECTION PRINTS AND CONVERTS THEM.

```
     IF(NEWIN.EQ.0) GO TO 20
     WRITE (6,1100)
     WRITE (6,1101)
     DO 10  J=1,NTOST
     IF(AFDRT(J)+OFDRT(J).EQ.0.0) GO TO 7
     IF(AFDRT(J).NE.0.) WRITE (6,1000)J,(XFD(I,J),I=1,6),AFDRT(J),
    .  AFDTEM(J)
     IF(OFDRT(J).NE.0.) WRITE (6,1001)CTBP,J,(YFD(I,J),I=1,6),
    .  OFDRT(J),OFDTEM(J)
```

CONVERSION TO MOLAR UNITS.

```
     XFD(2,J)=XFD(2,J)/238.
     YFD(2,J)=YFD(2,J)/238.
     XFD(3,J)=XFD(3,J)/239.
     YFD(3,J)=YFD(3,J)/239.
     XFD(4,J)=XFD(4,J)/239.
     YFD(4,J)=YFD(4,J)/239.
     XFD(5,J)=XFD(5,J)/238.
     YFD(5,J)=YFD(5,J)/238.
     DO 5 I=1,6
     AQ(I)=XFD(I,J)
     OR(I)=YFD(I,J)
   5 CONTINUE
     TEMP=OFDTEM(J)
```

SUBROUTINE MOLAL CALCULATES THE MOLAR TO MOLAL CONVERSION FACTORS.

```
     CALL MOLALP
     AFDRT(J)=AFDRT(J)/CONVA
     OFDRT(J)=OFDRT(J)/CONVO
     DO 8 I=1,NSOLU
     XFD(I,J)=XFD(I,J)*CONVA
     YFD(I,J)=YFD(I,J)*CONVO
```

```
 8 CONTINUE
 7 CONTINUE
10 CONTINUE


IF AN INITIAL PROFILE HAS BEEN GIVEN (IPRO = 1), THIS SECTION
CONVERTS THE CONCENTRATIONS AND INITIALIZES THE SETTLERS.

20 IF(IPRO.EQ.0) GO TO 40
   DO 30 J=1,NTOST
   X(2,J,2)=X(2,J,2)/238.
   Y(2,J,2)=Y(2,J,2)/238.
   X(3,J,2)=X(3,J,2)/239.
   Y(3,J,2)=Y(3,J,2)/239.
   X(4,J,2)=X(4,J,2)/239.
   Y(4,J,2)=Y(4,J,2)/239.
   X(5,J,2)=X(5,J,2)/238.
   Y(5,J,2)=Y(5,J,2)/238.
   DO 22 I=1,NSOLU
   AQ(I)=X(I,J,2)
   OR(I)=Y(I,J,2)
22 CONTINUE
   TEMP=TPROF(J,2)
   CALL MOLALP
   DO 25 I=1,6
   AQ(I)=AQ(I)*CONVA
   OR(I)=OR(I)*CONVO
   DO 25 L=1,2
   X(I,J,L)=AQ(I)
   Y(I,J,L)=OR(I)
   DO 25 K=1,3
   XS(J,K,I,L)=AQ(I)
   YS(J,K,I,L)=OR(I)
25 CONTINUE
30 CONTINUE
40 RETURN
000 FORMAT(14X,'AQUEOUS ',I3,7(4X,1PE10.3),3X,0PF7.1)
001 FORMAT(9X,2PF6.1,' % TBP ',I3,7(4X,1PE10.3),3X,0PF7.1)
100 FORMAT('-FEED & PRODUCT       STAGE  NITRIC ACID     U (VI)
   .  PU (IV)       PU (III)        U (IV)     NITRATE ION    FLOW RATE
   .     TEMP')
101 FORMAT(' STREAM DATA           NO.      (MOL/L)       (G/L)
   . (G/L)          (G/L)          (G/L)         (MOL/L)       (L/MIN)
   .      (C)')
    END
    SUBROUTINE MOLALP


SUBROUTINE MOLAL PROVIDES THE CONVERSION FACTORS (CONVA, CONVO) BETWEEN
MOLAR AND MOLAL UNITS.  AQ AND OR CONTAIN THE CONCENTRATIONS TO BE
CONVERTED.  TCONC SIGNALS THE UNITS OF THE CONCENTRATIONS BEING PASSED.
            TCONC = -1.0 FOR MOLAR CONCENTRATIONS
            TCONC =  1.0 FOR MOLAL CONCENTRATIONS

   COMMON/CONTRL/ NTOST,NSOLU,CTBP,NEWIN,NEWOUT,SPH,IPNCH,IFAST,IRXN,
  .  DTHETA,IVOLM,IVOLS,IPRO,TEMPI,ISOL(6),NSTR,ISTR,JSTR,CODUM(100)
   COMMON/MOLALC/ AQ(6),OR(6),TEMP,CONVA,CONVO,TCONC
   F=CTBP
   T=3.65145*F
   AQU = AQ(2)+AQ(5)
   AQPU = AQ(3)+AQ(4)
   ORU = OR(2)+OR(5)
   ORPU = OR(3)+OR(4)
```

```
      IF(TCONC.EQ.1.0) GO TO 5
      UOS=T/(2.0+0.092*T)
      PUOS=T/(2.0+0.18*T)
      HOS=T*(1.0-0.00609*(3.95-0.0144*TEMP)*(F**1.65))/(1.0+0.043*T)
      WO=(3.95-0.0144*TEMP)*(F**1.65)*(1.0-ORU/UOS-ORPU/PUOS-
     .     0.65*OR(1)/HOS)
      GO TO 10
    5 WO=(4.2-0.015*TEMP)*(F**1.69)*(T-2.0*ORU-2.0*ORPU-0.6*OR(1))/T
   10 CONVO= 1.0+TCONC*(0.097*ORU+0.139*ORPU+0.043*OR(1)+0.0174*WO)
      CONVO=ABS(CONVO)
      CONVO=CONVO**TCONC
      CONVA=0.0724*AQU+0.13*AQPU+0.0309*AQ(1)+0.031*AQ(6)
      CONVA=ABS(1.0+TCONC*CONVA)**TCONC
      RETURN
      END
      SUBROUTINE STAGES (NDIREC)
CCCCC


   SUBROUTINE STAGES IS NOT COMPLICATED, BUT MODIFICATIONS TO IT SHOULD BE
                    MADE WITH CARE.  IT EMPLOYS A FOURTH ORDER RUNGE-KUTTA
                    INTEGRATION OF THE DIFFERENTIAL EQUATIONS GOVERNING THE
                    CHANGES IN CONCENTRATIONS OF THE MIXERS.  CERTAIN VARIABLES
                    IN THE EQUATIONS ARE APPROXIMATED AS CONSTANTS FOR THE
                    TIME INTERVAL (AIN & OIN) AND DETERMINED BY RECTANGULAR
                    METHODS HALF THE TIMES, AND BY TRAPEZOIDAL METHODS THE
                    OTHER HALF (AS SPECIFIED BY NDIREC).
                    THE OTHER INTEGRATION METHODS ARE DIFFERENT.
                    IFAST=2 USES A SOLUTION TO THE FIRST ORDER LINEAR DIFFER-
                    ENTIAL EQUATION.
                    IFAST=1 USES A TRAPEZOIDAL APPROXIMATION TO THE
                    DIFFERENTIAL.
                       TEMPERATURE CHANGES ARE MADE BY A SIMPLE HEAT
                    BALANCE



  DESCRIPTION OF VARIABLES
     F(I)  AN EVALUATION OF THE DERIVATIVE IN THE INTEGRATION FOR COMPONENT I
     PHI(I)    WEIGHTED TOTAL OF THE VARIOUS F(I) COMPUTED
     TX(I)     TEST X FOR ITERATION BETWEEN THE CONCS AND THE DISTRIBUTION COEFF
     AIN IS THE SOLUTE COMING IN THE AQUEOUS STREAMS
     OIN IS THE SOLUTE COMING IN THE ORGANIC STREAMS
     DELTAX IS THE DIFFERENCE BETWEEN THE AIN VALUES AT T AND T+DELTA T
     DELTAY IS THE DIFFERENCE BETWEEN THE OIN VALUES AT T AND T+DELTA T
     ARY       USED IN SAME WAY AS TX, SORT OF
     DTRY      TRIAL DISTRIBUTION COEFFICIENTS
     NDIREC CHANGES THE STREAM THAT THE CALCULATIONS ARE FOLLOWING.  ONE TIME,
        THE CALCULATIONS WILL START WITH THE FIRST STAGE AND GO TO STAGE N
        (FOLLOWING THE AQUEOUS PHASE).  THE NEXT TIME IT WILL START WITH
        STAGE N, AND GO THE OTHER DIRECTION.


CCCCC
      COMMON/CONTRL/ NTOST,NSOLU,CTBP,NEWIN,NEWOUT,SPH,IPNCH,IFAST,IRXN,
     .  DTHETA,IVOLM,IVOLS,IPRO,TEMPI,ISOL(6),NSTR,ISTR,JSTR,CODUM(100),
     .  IPROCE
      COMMON/CONCS/ X(6,100,2),Y(6,100,2),XS(100,3,6,2),YS(100,3,6,2)
      COMMON/DISTRB/ TEMPC,ARY(6),DTRY(6)
      COMMON/FLOWS/ A(100),O(100),AT(100),OT(100)
      COMMON/MOLALC/ AQ(6),OR(6),TEMP,CONVA,CONVO,TCONC
      COMMON/STREAM/ XFD(6,100),YFD(6,100),AFDRT(100),OFDRT(100),
     .   AFDTEM(100),OFDTEM(100),ALVRT(100),OLVRT(100)
```

```
      COMMON/TEMPS/ TPROF(100,2),ATS(100,3,2),OTS(100,3,2)
      COMMON/VOLS/ AVOL(100),OVOL(100),ASVOL(100),OSVOL(100)
      COMMON/RXNS/ RXNTRM(6,100),AIN(6),OIN(6)
      COMMON/ELECT/ XP(6,100),XE(6,99),DX(6,99),FR,DIV,DT,
     .              PS,QS,MS,VC,BRU,BRPU,OC,OA,GAM
      DIMENSION F(6),PHI(6),TX(6),DELTAX(6),DELTAY(6)
      INTEGER Q,N,P,PS,QS,DIV
      CCD=0.001


 TCONC = 1.0 FOR THE VOLUME CONVERSIONS SINCE MOLAL CONCENTRATIONS ARE PASSED.

      TCONC=1.0
      DO 2 I=1,6
      F(I)=0.0
      TX(I)=0.0
      AQ(I)=0.0
      OR(I)=0.0
      ARY(I)=0.0
      AIN(I)=0.0
      OIN(I)=0.0
      PHI(I)=0.0
      DELTAX(I)=0.0
      DELTAY(I)=0.0
    2 CONTINUE


        MSCAN IS USED TO CYCLE THROUGH THE COLUMN CALCULATIONS TWICE FOR
        THE NEW INTEGRATION (IFAST=2)

      MSCAN=0
    3  CONTINUE
      DO 500 NSCAN=1,NTOST
      J=NSCAN
      IF(NDIREC.LT.0) J=NTOST-NSCAN+1
      JJ=J-1
      JJJ=J+1
      CODUM(J)=0.0


     CALL SUBROUTINE ELECTROCELL WHEN IRXN .GE. 4 BETWEEN STAGE PS & PS+1
     AND QS & QS+1, WHERE ELECTROCELLS ARE CONNECTED

      IF((IRXN.LT.4).OR.(J.NE.PS+1)) GO TO 470
        MS=PS
        CALL ELECTRO
  470 CONTINUE
      IF((IRXN.LT.4).OR.(J.NE.QS+1)) GO TO 471
        MS=QS
        CALL ELECTRO


 ALL OF THIS 'DO 7' LOOP IS CONCERNED WITH ACCUMULATING THE APPROXIMATION
      FOR THE INPUT CONCENTRATIONS

  471 DO 7 I=1,NSOLU
      IF(ISOL(I).EQ.0) GO TO 7
        DELTAX(I)=0.0
        DELTAY(I)=0.0


 ADDING THE SOLUTES IN THE FEED STREAMS
```

```
      AIN(I)=AFDRT(J)*XFD(I,J)
      OIN(I)=OFDRT(J)*YFD(I,J)


ADDING THE AQUEOUS SOLUTE FROM THE PRECEEDING STAGE.  IF NDIREC IS
POSITIVE, THE PRECEEDING STAGE HAS A VALUE FOR TWO POINTS IN TIME,
SO A TRAPEZOIDAL APPROXIMATION IS USED.

      IF(J.EQ.1) GO TO 4
      AIN(I)=AIN(I)+A(JJ)*XS(JJ,3,I,1)
      DELTAX(I)=(XS(JJ,3,I,2)-XS(JJ,3,I,1))*A(JJ)
      IF(IFAST.NE.2) AIN(I)=AIN(I)+(XS(JJ,3,I,2)-XS(JJ,3,I,1))*A(JJ)/2.0
    4 CONTINUE


A SIMILAR PROCESS FOR THE ORGANIC SOLUTE FROM THE SUCCEEDING STAGE.

      IF(J.EQ.NTOST) GO TO 5
      OIN(I)=OIN(I)+O(JJJ)*YS(JJJ,3,I,1)
      DELTAY(I)=(YS(JJJ,3,I,2)-YS(JJJ,3,I,1))*O(JJJ)
      IF(IFAST.NE.2) OIN(I)=OIN(I)
     .     +(YS(JJJ,3,I,2)-YS(JJJ,3,I,1))*O(JJJ)/2.0
    5 CONTINUE

IF NSTR = 1 AN ORGANIC STREAM MUST BE TREATED IN A SPECIAL MANNER.

      IF(NSTR.NE.1.OR.(J.NE.JSTR.AND.J.NE.ISTR-1)) GO TO 6
      IF(J.EQ.ISTR-1) OIN(I)=OFDRT(J)*YFD(I,J)
      IF(J.EQ.ISTR-1) DELTAY(I)=0.0
      IF(J.EQ.JSTR) OIN(I)=OIN(I)+O(ISTR)*YS(ISTR,3,I,1)
      IF(J.EQ.JSTR) DELTAY(I)=DELTAY(I)+(YS(ISTR,3,I,2)-YS(ISTR,3,I,1)
     .     )*O(ISTR)
      IF(J.EQ.JSTR.AND.IFAST.NE.2) OIN(I)=OIN(I)+(YS(ISTR,3,I,2)
     .     -YS(ISTR,3,I,1))*O(ISTR)/2.0
    6 CONTINUE

THE PROGRAM WILL NOT DEAL WITH SOLUTE FLOWS SMALLER THAN THIS
THRESHOLD LEVEL.

      IF(ABS(AIN(I)).LT.1.0E-15) AIN(I)=0.0
      IF(ABS(OIN(I)).LT.1.0E-15) OIN(I)=0.0
      TX(I)=X(I,J,2)
      ARY(I)=X(I,J,2)
      AQ(I)=X(I,J,2)
      OR(I)=Y(I,J,2)
    7 CONTINUE

GETTING MOLAL VOLUMES
      SOLUTE FREE VOLUMES ARE REQUIRED TO CORRECT THE UNITS OF THE VOLUMES
      FROM REAL VOLUME UNITS TO MOLAL UNITS IN ORDER TO MAKE THE V*X VALID

      TEMP=TPROF(J,2)
      CALL MOLALP
      AQVOL=AVOL(J)/CONVA
      ORVOL=OVOL(J)/CONVO
      AQSVOL=ASVOL(J)/CONVA
      ORSVOL=OSVOL(J)/CONVO

THE HEAT BALANCES ARE DONE IN A WAY SIMILAR TO THE CONCENTRATIONS

      AHTIN=AFDRT(J)*AFDTEM(J)
      OHTIN=OFDRT(J)*OFDTEM(J)
      IF(J.NE.1) AHTIN=AHTIN+A(JJ)*ATS(JJ,3,1)
```

```
      IF(J.NE.1) AHTIN=AHTIN+(ATS(JJ,3,2)-ATS(JJ,3,1))*A(JJ)/2.
      IF(J.NE.NTOST) OHTIN=OHTIN+O(JJJ)*OTS(JJJ,3,1)
      IF(J.NE.NTOST) OHTIN=OHTIN+(OTS(JJJ,3,2)-OTS(JJJ,3,1))*O(JJJ)/2.
      IF(NSTR.EQ.1.AND.J.EQ.ISTR-1) OHTIN=OFDRT(J)*OFDTEM(J)
      IF(NSTR.EQ.1.AND.J.EQ.JSTR) OHTIN=OHTIN+O(ISTR)*OTS(ISTR,3,1)
      IF(NSTR.EQ.1.AND.J.EQ.JSTR)
     .   OHTIN=OHTIN+(OTS(ISTR,3,2)-OTS(ISTR,3,1))*O(ISTR)/2.
      TPROF(J,2)=TPROF(J,1)+DTHETA*(AHTIN+SPH*OHTIN-(AT(J)+SPH*OT(J))
     .   *TPROF(J,1))/(AQVOL+SPH*ORVOL)
      TEMPC=TPROF(J,2)


      THIS BRANCH IS FOR THE FASTER INTEGRATION METHOD.

      IMCHEM=0
      IF(IFAST.EQ.1) GO TO 150
      IF(IFAST.EQ.2) GO TO 135
CC


      THIS IS THE START OF THE RUNGE-KUTTA INTEGRATION


CC
      MCK=0

      GET THE DISTRIBUTION COEFF FROM THE PREVIOUS TIME

      CALL UCORPU
   10 DO 20 I=1,NSOLU

      CALCULATE THE FIRST DERIVATIVE

      F(I)=(AIN(I)+OIN(I)-(AT(J)+OT(J)*DTRY(I))*X(I,J,1))/
     .     (AQVOL+ORVOL*DTRY(I))

      INCREASE X(I) BY DELTA X /2

   20 ARY(I)=X(I,J,1)+DTHETA*F(I)/2.
      MCK=MCK+1

      GETTING A NEW DTRY WITH THE SLIGHTLY LARGER X'S

      CALL UCORPU

      ITERATING TO GET MATCHING DTRY'S AND X'S (ARY'S)

      MDEX=0
      DO 30 I=1,NSOLU
      IF(ABS(ARY(I)).LT.1.0E-6) GO TO 30
      DIF=ABS((TX(I)-ARY(I))/ARY(I))
      IF(DIF.GT.CCD) MDEX=1
   30 TX(I)=ARY(I)
      IF(MDEX.EQ.1.AND.MCK.LT.5) GO TO 10
      DO 40 I=1,NSOLU
      X(I,J,2)=ARY(I)
   40 PHI(I)=F(I)

   THIS (OR SIMILAR) PROCESS IS REPEATED WHILE ACCUMULATING PHI'S FOR THE
      FINAL DETERMINATION OF X AT THE NEW TIME
   45 DO 50 I=1,NSOLU
      F(I)=(AIN(I)+OIN(I)-(AT(J)+OT(J)*DTRY(I))*X(I,J,2))/
     .     (AQVOL+ORVOL*DTRY(I))
```

```
 50 ARY(I)=X(I,J,1)+DTHETA*F(I)/2.
    MCK=MCK+1
    CALL UCORPU
    MDEX=0
    DO 60 I=1,NSOLU
    IF(ABS(ARY(I)).LT.1.0E-6) GO TO 60
    DIF=ABS((TX(I)-ARY(I))/ARY(I))
    IF(DIF.GT.CCD)MDEX=1
 60 TX(I)=ARY(I)
    IF(MDEX.EQ.1.AND.MCK.LT.10) GO TO 45
    DO 70 I=1,NSOLU
    X(I,J,2)=ARY(I)
 70 PHI(I)=PHI(I)+2.*F(I)
 75 DO 80 I=1,NSOLU
    F(I)=(AIN(I)+OIN(I)-(AT(J)+OT(J)*DTRY(I))*X(I,J,2))/
   .      (AQVOL+ORVOL*DTRY(I))
 80 ARY(I)=X(I,J,1)+DTHETA*F(I)
    MCK=MCK+1
    CALL UCORPU
    MDEX=0
    DO 90 I=1,NSOLU
    IF(ABS(ARY(I)).LT.1.0E-6) GO TO 90
    DIF=ABS((TX(I)-ARY(I))/ARY(I))
    IF(DIF.GT.CCD) MDEX=1
 90 TX(I)=ARY(I)
    IF(MDEX.EQ.1.AND.MCK.LT.15) GO TO 75
    DO 100 I=1,NSOLU
    X(I,J,2)=ARY(I)
100 PHI(I)=PHI(I)+2.*F(I)
105 DO 110 I=1,NSOLU
    F(I)=(AIN(I)+OIN(I)-(AT(J)+OT(J)*DTRY(I))*X(I,J,2))/
   .      (AQVOL+ORVOL*DTRY(I))
110 ARY(I)=X(I,J,1)+(PHI(I)+F(I))*DTHETA/6.
    MCK=MCK+1
    CALL UCORPU
    MDEX=0
    DO 120 I=1,NSOLU
    IF(ABS(ARY(I)).LT.1.0E-6) GO TO 120
    DIF=ABS((TX(I)-ARY(I))/ARY(I))
    IF(DIF.GT.CCD) MDEX=1
120 TX(I)=ARY(I)
    IF(MDEX.EQ.1.AND.MCK.LT.20) GO TO 105
    DO 130 I=1,NSOLU
    IF(ARY(I).LE.1.E-20) ARY(I)=0.0
    Y(I,J,2)=DTRY(I)*ARY(I)
130 X(I,J,2)=ARY(I)
135  CONTINUE
     MCK=0
136  MDEX=0
     CALL UCORPU
       DO 140 I=1,NSOLU
     WGHT=DTHETA*(AT(J)+DTRY(I)*OT(J))/(AQVOL+DTRY(I)*ORVOL)
     ARY(I)=X(I,J,1)*EXP(-WGHT)+(AIN(I)+OIN(I)+RXNTRM(I,J))
   .      *(1.0-EXP(-WGHT))/(AT(J)+DTRY(I)*OT(J))+(DELTAX(I)+DELTAY(I))
   .      *(EXP(-WGHT)+WGHT-1.0)/(WGHT*(AT(J)+DTRY(I)*OT(J)))
     DIF=0.0
     IF(ABS(ARY(I)).GE.1.0E-6) DIF=ABS((X(I,J,2)-ARY(I))/ARY(I))
     IF(DIF.GT.CCD) MDEX=1
     IF (ARY(I).LT.1.0E-10) ARY(I)=0.0
     X(I,J,2)=ARY(I)
     Y(I,J,2)=ARY(I)*DTRY(I)
140  CONTINUE
     MCK=MCK+1
```

```
      IF(MDEX.EQ.1.AND.MCK.LT.15) GO TO 136
      IF(IMCHEM.EQ.1.OR.IPROCE.EQ.1) GO TO 185
      IF(IRXN.EQ.0.OR.IRXN.EQ.4) GO TO 185
      IMCHEM=IMCHEM+1
      CALL MCHEMP(J,AQVOL,ORVOL)
      GO TO 135

  THIS COMPLETES THE INTEGRATION.  ARY(I) (FROM STM 110) BECOMES THE NEW
     AQUEOUS CONCENTRATIONS
     AND NOW THE SETTLER VALUES ARE CALCULATED WITH A TRAPEZOIDAL METHOD

     GO TO 180
C


     THIS IS THE FASTER INTEGRATION METHOD.  IT IS VERY SIMILAR TO ONE
     OF THE SEGMENTS OF THE RUNGE-KUTTA INTEGRATION.  IT USES A SIMPLER
     FINITE DIFFERENCE METHOD TO EVALUATE THE CONCENTRATIONS IN THE
     MIXERS.  ALL OTHER CALCULATIONS ARE DONE IN THE SAME WAY AS FOR
     THE RUNGE-KUTTA INTEGRATION

C
 150 MCK=0
 160 MDEX=0
     CALL UCORPU
     DO 170 I=1,NSOLU
     IF(ISOL(I).EQ.0) GO TO 170
     DVOLS=AQVOL+DTRY(I)*ORVOL
     DFLOS=AT(J)+DTRY(I)*OT(J)
     X(I,J,2)=(AQVOL*X(I,J,1)+ORVOL*Y(I,J,1)+DTHETA*(AIN(I)+OIN(I)-
    .      (AT(J)*X(I,J,1)+OT(J)*Y(I,J,1))/2.))/(DVOLS+DTHETA*DFLOS/2.)
     IF(X(I,J,2).LE.1.E-20) X(I,J,2)=0.0
     Y(I,J,2)=X(I,J,2)*DTRY(I)
     IF(ABS(X(I,J,2)).LT.1.E-09) GO TO 165
     DIF=ABS((X(I,J,2)-ARY(I))/X(I,J,2))
     IF(DIF.GE.CCD) MDEX=1
 165 ARY(I)=X(I,J,2)
 170 CONTINUE
     MCK=MCK+1
     IF(MDEX.EQ.1.AND.MCK.LE.20) GO TO 160

 180 FACTA=0.0
     FACTO=0.0


  SUBROUTINE MCHEMP DEALS WITH ANY CHEMICAL REACTIONS OCCURING IN THE MIXERS.

     IF(IRXN.EQ.0.OR.IRXN.EQ.4) GO TO 185
     CALL MCHEMP(J,AQVOL,ORVOL)
 185 CONTINUE


      FACTA=150.0
      FACTO=150.0
     IF(AQSVOL.GT.0.0) FACTA=3.0*AT(J)*DTHETA/AQSVOL
     IF(ORSVOL.GT.0.0) FACTO=3.0*OT(J)*DTHETA/ORSVOL
     ATS(J,1,2)=ATS(J,1,1)*EXP(-FACTA)+TPROF(J,1)*(1.0-EXP(-FACTA))
    .      +(TPROF(J,2)-TPROF(J,1))*(EXP(-FACTA)+FACTA-1.0)/FACTA
     OTS(J,1,2)=OTS(J,1,1)*EXP(-FACTO)+TPROF(J,1)*(1.0-EXP(-FACTO))
    .      +(TPROF(J,2)-TPROF(J,1))*(EXP(-FACTO)+FACTO-1.0)/FACTO
     DO 200 I=1,NSOLU
     IF(ISOL(I).EQ.0) GO TO 200
     CNGINV=0.0
```

```
      IF(X(I,J,2)+Y(I,J,2).GT.1.0E-6) CNGINV=(AVOL(J)*(X(I,J,2)-X(I,J,1)
     .)+OVOL(J)*(Y(I,J,2)-Y(I,J,1)))/(DTHETA*(AVOL(J)*X(I,J,2)+OVOL(J)*
     .Y(I,J,2)))
      CNGINV=100*ABS(CNGINV)
      CODUM(J)=AMAX1(CODUM(J),CNGINV)
      XS(J,1,I,2)=XS(J,1,I,1)*EXP(-FACTA)+X(I,J,1)*(1.0-EXP(-FACTA))
     .     +(X(I,J,2)-X(I,J,1))*(EXP(-FACTA)+FACTA-1.0)/FACTA
      YS(J,1,I,2)=YS(J,1,I,1)*EXP(-FACTO)+Y(I,J,1)*(1.0-EXP(-FACTO))
     .     +(Y(I,J,2)-Y(I,J,1))*(EXP(-FACTO)+FACTO-1.0)/FACTO
  200 CONTINUE
      DO 300 KZ=2,3
      KZB=KZ-1
      ATS(J,KZ,2)=ATS(J,KZ,1)*EXP(-FACTA)+ATS(J,KZB,1)*(1.0-EXP(-FACTA))
     .     +(ATS(J,KZB,2)-ATS(J,KZB,1))*(EXP(-FACTA)+FACTA-1.0)/FACTA
      OTS(J,KZ,2)=OTS(J,KZ,1)*EXP(-FACTO)+OTS(J,KZB,1)*(1.0-EXP(-FACTO))
     .     +(OTS(J,KZB,2)-OTS(J,KZB,1))*(EXP(-FACTO)+FACTO-1.0)/FACTO
      DO 300 I=1,NSOLU
      XS(J,KZ,I,2)=XS(J,KZ,I,1)*EXP(-FACTA)+XS(J,KZB,I,1)
     .     *(1.0-EXP(-FACTA))+(XS(J,KZB,I,2)-XS(J,KZB,I,1))
     .     *(EXP(-FACTA)+FACTA-1.0)/FACTA
      YS(J,KZ,I,2)=YS(J,KZ,I,1)*EXP(-FACTO)+YS(J,KZB,I,1)
     .     *(1.0-EXP(-FACTO))+(YS(J,KZB,I,2)-YS(J,KZB,I,1))
     .     *(EXP(-FACTO)+FACTO-1.0)/FACTO
  300 CONTINUE
      IF(IRXN.EQ.0.OR.IRXN.EQ.4) GO TO 500


 SUBROUTINE SCHEMP DEALS WITH ANY CHEMICAL REACTIONS OCCURING IN THE
 SETTLERS.  THESE REACTIONS ARE SIMPLER SINCE NO PHASE TRANSFER IS
 POSSIBLE.

      CALL SCHEMP(J,AQSVOL,ORSVOL)
  500 CONTINUE
      NDIREC=-NDIREC
      MSCAN=MSCAN+1
      IF(IFAST.EQ.2.AND.MSCAN.LT.2) GO TO 3
      IF(IFAST.NE.2) NDIREC=-NDIREC
      RETURN
      END


      SUBROUTINE UCORPU
      COMMON/CONTRL/ NTOST,NSOLU,CTBP,NEWIN,NEWOUT,SPH,IPNCH,IFAST,IRXN,
     .  DTHETA,IVOLM,IVOLS,IPRO,TEMPI,ISOL(6),NSTR,ISTR,JSTR,CODUM(100),
     .  IPROCE
      COMMON/DISTRB/ TEMPC,ARY(6),DTRY(6)

 MODIFIED TO CALCULATE DISTRIBUTION COEFFICIENTS FOR U(IV) & PU(III)

 UCOR RETURNS A VALUE OF THE DISTRIBUTION COEFFICIENT = Y/X FOR EACH
   VALUE OF X TRIED

      PUTOT=ARY(3)+ARY(4)
      UTOT = ARY(2)+ARY(5)
      F=CTBP
      HAM = ARY(1)
      UAM = ARY(2)
      PUAM = ARY(3)
      SNITR = ARY(6)
      IF(UAM.LT.0) UAM = 0.
      IF(PUAM.LT.0) PUAM = 0.
      IF(HAM.LT.0) HAM = 0.
      IF(SNITR.LT.0) SNITR = 0.
```

```
      PUIII = PUTOT - PUAM
      UIV = UTOT - UAM
      IF(PUIII.LT.0) PUIII = 0.
      IF(UIV.LT.0) UIV = 0.
      TNM = HAM + 2.*UAM + 2.*PUAM + SNITR  + 2.*PUIII + 3.*UIV
      IF (TNM.EQ.0.0) TNM = 1.0
      TEMPRK = 1000./(TEMPC + 273.16)
      DRT = TEMPRK - 3.3539
      UK = 3.7*TNM**1.57 + 1.4*TNM**3.9 + 0.011*TNM**7.3
      UK = UK*1.13*(4.*F**(-0.17) - 3.)
      PUK = UK*(.135 + .55*F**1.25 + .010*TNM**2)
      UIVK = UK*(0.0220 + 0.000981*TNM**2)
      PIIIK = 0.04*TNM**1.8+0.000156*F*TNM**7
      HK1 = 0.135*TNM**0.82 + 0.0052*TNM**3.44
      IF (F.LT.1.) HK1 = HK1*(1. - 0.54*EXP(-15.*F))

      IF (TEMPC.NE.25.) UK = UK*EXP(2.5*DRT)
      IF (TEMPC.NE.25.) PUK = PUK*EXP(-.8*DRT)
      IF (TEMPC.NE.25.) UIVK = UIVK*EXP(-.2*DRT)
      IF (TEMPC.NE.25.) HK1 = HK1*EXP(0.34*DRT)

      HK2 = HK1
      A = 2.*(UK*UAM + PUK*PUAM + HK2*HAM + UIVK*UIV + PIIIK*PUIII)
      B = HK1*HAM + 1.
      C=-3.65145*CTBP
      IF(A.GE.1.E-6) GO TO 10
      TF=-C/B
      GO TO 15
10    CONTINUE
      TF = (-B + SQRT(B**2 -4.*A*C))/(2.*A)
15    CONTINUE
      DH1 = HK1*TF
      DH2 = HK2*TF**2
      DH = DH1 + DH2
      DU = UK*TF**2
      DPU = PUK*TF**2
      DPUT = PIIIK*TF**2
      DUF = UIVK*TF**2
      DENOM=0.598183+0.0131237*TEMPC
      DENOM=1.0-.43223+.013118*TEMPC+.743579*UAM-.17583*UAM*HAM
      DPU=DPU/DENOM

      DTRY(1) = DH
      DTRY(2) = DU
      DTRY(3) = DPU
      DTRY(4) = DPUT
      DTRY(5) = DUF
      DTRY(6) = 0.0

20    RETURN
      END
      SUBROUTINE MCHEMP (J,AQVOL,ORVOL)
```

SUBROUTINE MCHEM HANDLES THE CHEMICAL REACTIONS IN THE SYSTEM. PRESENTLY, INSTANTANEOUS REDUCTION OF PLUTONIUM, REDUCTION BY U(IV), AND REDUCTION BY HYDROXYLAMINE ARE AVAILABLE AS REACTIONS. THE SUBROUTINE CAN EASILY BE ADAPTED TO CONSIDER ANY INTEGRATED RATE EQUATION, WITH ANY DESIRED STOICHIOMETRY.
THE SUBROUTINE ASSUMES THE REACTION IS TOTALLY IN THE AQUEOUS PHASE, BUT THAT THE SOLUTE IN THE ORGANIC PHASE ALSO AIDS IN MAINTAINING THE AQUEOUS CONCENTRATION. THE INTEGRATED RATE EQUATION DETERMINES THE EXTENT OF REACTION. THE ROUTINE THEN SPLITS THE RESULTING SOLUTES BETWEEN THE PHASES.

```
IRXN INDICATES WHICH REACTION RATE IS TO BE USED.
SOLAMT IS THE TOTAL AMOUNT OF SOLUTE IN THE MIXER
SOLVOL IS A PSEUDO-VOLUME SO THAT THE SOLUTE IS ENTIRELY IN THE AQUEOUS PHASE
RX IS THE AQUEOUS COMPOSITION USED TO DETERMINE THE REACTION RATE
RXNAMT IS THE AMOUNT OF REACTED SOLUTE USING SOME COMPONENT AS A BASIS
RK IS A REACTION RATE CONSTANT
EXTENT = THE EXTENT OF REACTION, BASED ON THE FRACTION OF
          SOME COMPONENT CONSUMED BY THE REACTION.


   COMMON/CONTRL/ NTOST,NSOLU,CTBP,NEWIN,NEWOUT,SPH,IPNCH,IFAST,IRXN,
  .  DTHETA,IVOLM,IVOLS,IPRO,TEMPI,ISOL(6),NSTR,ISTR,JSTR,CODUM(100),
  .  IPROCE
   COMMON/CONCS/ X(6,100,2),Y(6,100,2),XS(100,3,6,2),YS(100,3,6,2)
   COMMON/DISTRB/ TEMPC,ARY(6),DTRY(6)
   COMMON/TEMPS/ TPROF(100,2),ATS(100,3,2),OTS(100,3,2)
   COMMON/RXNS/ RXNTRM(6,100),AIN(6),OIN(6)
   DIMENSION RX(6),SOLAMT(6),SOLVOL(6)
    REAL STOIC(6,3)/0.0,0.0,-1.0,1.0,-1.0,0.0,
   .                2.0,0.5,-1.0,1.0,-0.5,0.0,
   .                2.0,0.0,-1.0,1.0,-1.0,0.0/
     DIMENSION STOIC(6,7)
     DO 1 ISTOIC=1,6
     DO 1 JSTOIC=1,7
     STOIC(ISTOIC,JSTOIC)=0.0
     CONTINUE
     STOIC(3,1)=-1.0
     STOIC(4,1)=1.0
     STOIC(5,1)=-1.0

     STOIC(1,2)=2.0
     STOIC(2,2)=0.5
     STOIC(3,2)=-1.0
     STOIC(4,2)=1.0
     STOIC(5,2)=-0.5

     STOIC(1,3)=2.0
     STOIC(3,3)=-1.0
     STOIC(4,3)=1.0
     STOIC(5,3)=-1.0

     STOIC(1,5)=2.0
     STOIC(3,5)=-1.0
     STOIC(4,5)=1.0
     STOIC(5,5)=-1.0

     STOIC(1,6)=2.0
     STOIC(2,6)=0.5
     STOIC(3,6)=-1.0
     STOIC(4,6)=1.0
     STOIC(5,6)=-0.5

     STOIC(3,7)=-1.0
     STOIC(4,7)=1.0
     STOIC(5,7)=-1.0

   IF(ISOL(5).EQ.0) GO TO 300
   DO 10 I=1,6
   SOLAMT(I)=AQVOL*X(I,J,2)+ORVOL*Y(I,J,2)
   SOLVOL(I)=AQVOL+DTRY(I)*ORVOL
   RX(I)=X(I,J,2)
10 CONTINUE
```

```
   GO TO (90,80,70,60,50,40,30,20),IRXN
20 CONTINUE
30 CONTINUE
   GO TO 90
40 CONTINUE
   GO TO 80
50 CONTINUE
   GO TO 70
60 CONTINUE
   GO TO 130
70 CONTINUE


IRXN = 3  REACTION BETWEEN PU(IV) AND HYDROXYLAMINE
   FOR = RATIO OF PU(IV) TO REDUCTANT
   RK = RATE CONSTANT
   EXTMAX = MAXIMUM EXTENT OF REACTION (BASED ON PU (IV))
   EXTINC = INCREMENT IN SEARCH FOR EXTENT
   AMTINT = THE INTEGRATED CHANGE IN EXTENT (EQUAL TO RK)

   IF(RX(1).LT.1.0E-10.OR.RX(3).LT.1.0E-10.OR.RX(5).LT.1.0E-10)
  .     GO TO 90
   FOR=SOLAMT(3)/SOLAMT(5)
   TOTNIT=RX(1)+2.*RX(2)+4.*RX(3)+3.*RX(4)+RX(6)+0.33
   RK=1.74*EXP(31000./1.987*(1.0/303.16-1.0/(273.16+TPROF(J,2))))
   RK=RK*RX(3)*DTHETA*(RX(5)/(RX(1)*RX(1)*TOTNIT))**2.0
   EXTMAX=AMIN1(1.0,1.0/FOR)


   THIS IS A BINARY SEARCH FOR THE CORRECT EXTENT OF THE REACTION

   EXTINC=0.25*EXTMAX
   EXTENT=0.5*EXTMAX
   B=SOLAMT(3)/SOLVOL(4)
   A=RX(4)+B
   AA=A*A
   BB=A*B
   CC=B*B
   DO 78 I=1,10
   A=1.0/(1.0-EXTENT)
   IF(FOR.GT.0.01) GO TO 72


   THIS INTEGRATED FORM ASSUMES A LARGE EXCESS OF THE REDUCTANT

   AMTINT=AA*A*EXTENT-2.0*BB*ALOG(A)+CC*EXTENT
   GO TO 76
72 IF(ABS(1.0-FOR).LT.0.01) GO TO 74


   THIS INTEGRATED RATE EQUATION IS THE GENERAL CASE

   B=1.0-FOR*EXTENT
   C=1.0/(1.0-FOR)
   AMTINT=(FOR-A*EXTENT-FOR/B+2.0*C*FOR*ALOG(A*B))*AA*C*C
   AMTINT=AMTINT-2.0*BB*C*(FOR*EXTENT/B-C*ALOG(A*B))-CC*EXTENT/B
   AMTINT=-AMTINT
   GO TO 76


   THIS INTEGRATED FORM ASSUMES A STOICHIOMETRIC AMOUNT OF REDUCTANT
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```
 74 AMTINT=AA*(A*A*A-1.0)/3.0-BB*(A*A-1.0)+CC*(A-1.0)
 76 CONTINUE
    IF(AMTINT.GT.RK) EXTENT=EXTENT-EXTINC
    IF(AMTINT.LT.RK) EXTENT=EXTENT+EXTINC
    EXTINC=EXTINC/2.0
 78 CONTINUE
    RXNAMT=EXTENT*SOLAMT(3)
    GO TO 100
 80 CONTINUE


 IRXN = 2  REACTION BETWEEN PU (IV) AND U (IV)
    RK = RATE CONSTANT
    RCU = RATIO OF PU (IV) TO REDUCTANT
    EXTMAX = MAXIMUM EXTENT BASED ON PU (IV) AND A TEN SECOND HALF TIME

    IF(RX(3).LT.1.0E-10.OR.RX(5).LT.1.0E-10) GO TO 90
    EXTMAX=AMIN1(1.0,1.0-0.5**(6.0*DTHETA),2.0*SOLAMT(5)/SOLAMT(3))
    RK=170.0
    RK=RK*DTHETA*RX(5)
    RCU=SOLAMT(3)/(2.0*SOLAMT(5))
    RHO=RX(1)*RX(1)
    REXP=1.020
    IF(RK*(1.0-RCU).LT.100.0*RHO) REXP=EXP(RK*(1.0-RCU)/RHO)
    EXTENT=RK/(RK+RHO)
    IF(ABS(1.0-RCU).GT.0.05) EXTENT=(1.0-REXP)/(RCU-REXP)
    EXTENT=AMIN1(EXTENT,EXTMAX)
    RXNAMT=SOLAMT(3)*EXTENT
    GO TO 100
 90 CONTINUE


 IRXN = 1  INSTANTANEOUS REDUCTION OF PU (IV)
    THIS MECHANISM IS USED BY ALL REDUCTION REACTIONS WHEN THE
    CONCENTRATIONS FALL BELOW A THRESHHOLD LEVEL

    RXNAMT=AMIN1(-SOLAMT(3)/STOIC(3,IRXN),-SOLAMT(5)/STOIC(5,IRXN))
    IF(IFAST.EQ.2) RXNAMT=DTHETA*AMIN1(-(AIN(3)+OIN(3))
   .    /STOIC(3,IRXN),-(AIN(5)+OIN(5))/STOIC(5,IRXN))
100 DO 110 I=1,6
    SOLAMT(I)=SOLAMT(I)+RXNAMT*STOIC(I,IRXN)
    IF(SOLAMT(I).LT.0.0) SOLAMT(I)=0.0
    ARY(I)=SOLAMT(I)/SOLVOL(I)
    RXNTRM(I,J)=RXNAMT*STOIC(I,IRXN)/DTHETA
    IF(RXNTRM(I,J).NE.0.0) ISOL(I)=1
    IF(ISOL(I).EQ.1.AND.NSOLU.LT.I) NSOLU=I
110 CONTINUE
    IF(IFAST.EQ.2) GO TO 130


 DIVIDING THE RESULTING SOLUTES BETWEEN THE PHASES

    DO 125 ITRY=1,5
    ICK=0
    CALL UCORPU
    DO 120 I=1,NSOLU
    X(I,J,2)=SOLAMT(I)/(AQVOL+DTRY(I)*ORVOL)
    IF(ABS(X(I,J,2)-ARY(I)).GT.0.0001*X(I,J,2)) ICK=1
    ARY(I)=X(I,J,2)
    IF(X(I,J,2).LE.1.E-20) X(I,J,2)=0.0
    Y(I,J,2)=X(I,J,2)*DTRY(I)
120 CONTINUE
    IF(ICK.EQ.0) GO TO 130
```

```
125 CONTINUE
130 CONTINUE
300 RETURN
    END
    SUBROUTINE SCHEMP (J,AQSVOL,ORSVOL)


 SUBROUTINE SCHEM WORKS SIMILAR TO MCHEM, BUT THE PHASES ARE KEPT SEPARATE
 AT ALL TIMES.

 IRXN INDICATES WHICH REACTION RATE IS TO BE USED.
 RK IS A REACTION RATE CONSTANT
 EXTENT = THE EXTENT OF REACTION, BASED ON THE FRACTION OF
           SOME COMPONENT CONSUMED BY THE REACTION.



    COMMON/CONTRL/ NTOST,NSOLU,CTBP,NEWIN,NEWOUT,SPH,IPNCH,IFAST,IRXN,
   .  DTHETA,IVOLM,IVOLS,IPRO,TEMPI,ISOL(6),NSTR,ISTR,JSTR,CODUM(100),
   .  IPROCE
    COMMON/CONCS/ X(6,100,2),Y(6,100,2),XS(100,3,6,2),YS(100,3,6,2)
    COMMON/TEMPS/ TPROF(100,2),ATS(100,3,2),OTS(100,3,2)
     REAL STOICX(6,3)/0.0,0.0,-1.0,1.0,-1.0,0.0,
    .                 2.0,0.5,-1.0,1.0,-0.5,0.0,
    .                 2.0,0.0,-1.0,1.0,-1.0,0.0/
     REAL STOICY(6,3)/0.0,0.0,0.0,0.0,0.0,0.0,12*0.0/
    DIMENSION RX(6),RY(6),STOICX(6,7),STOICY(6,7)
    IF(ISOL(5).EQ.0) GO TO 300
    DO 150 KZ=1,3
    DO 9 I=1,6
    RX(I)=XS(J,KZ,I,2)
    RY(I)=YS(J,KZ,I,2)
  9 CONTINUE

      DO 10 ISTOIC=1,6
      DO 10 JSTOIC=1,7
      STOICX(ISTOIC,JSTOIC)=0.0
      STOICY(ISTOIC,JSTOIC)=0.0
 10   CONTINUE

      STOICX(3,1)=-1.0
      STOICX(4,1)=1.0
      STOICX(5,1)=-1.0

      STOICX(1,2)=2.0
      STOICX(2,2)=0.5
      STOICX(3,2)=-1.0
      STOICX(4,2)=1.0
      STOICX(5,2)=-0.5

      STOICX(1,3)=2.0
      STOICX(3,3)=-1.0
      STOICX(4,3)=1.0
      STOICX(5,3)=-1.0

      STOICX(1,5)=2.0
      STOICX(3,5)=-1.0
      STOICX(4,5)=1.0
      STOICX(5,5)=-1.0

      STOICX(1,6)=2.0
      STOICX(2,6)=0.5
      STOICX(3,6)=-1.0
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```
      STOICX(4,6)=1.0
      STOICX(5,6)=-0.5

      STOICX(3,7)=-1.0
      STOICX(4,7)=1.0
      STOICX(5,7)=-1.0

   GO TO (90,80,70,60,50,40,30,20),IRXN
20 CONTINUE
30 CONTINUE
   GO TO 90
40 CONTINUE
   GO TO 80
50 CONTINUE
   GO TO 70
60 CONTINUE
   GO TO 150
70 CONTINUE


IRXN = 3  REACTION BETWEEN PU(IV) AND HYDROXYLAMINE
   FOR = RATIO OF PU(IV) TO REDUCTANT
   RK = RATE CONSTANT
   EXTMAX = MAXIMUM EXTENT OF REACTION (BASED ON PU (IV))
   EXTINC = INCREMENT IN SEARCH FOR EXTENT
   AMTINT = THE INTEGRATED CHANGE IN EXTENT (EQUAL TO RK)

   IF(RX(1).LT.1.0E-10.OR.RX(3).LT.1.0E-10.OR.RX(5).LT.1.0E-10)
  .      GO TO 90
   FOR=RX(3)/RX(5)
   TOTNIT=RX(1)+2.*RX(2)+4.*RX(3)+3.*RX(4)+RX(6)+0.33
   RK=1.74*EXP(31000./1.987*(1.0/303.16-1.0/(273.16+TPROF(J,2))))
   RK=RK*RX(3)*DTHETA*(RX(5)/(RX(1)*RX(1)*TOTNIT))**2.0
   EXTMAX=AMIN1(1.0,1.0/FOR)


   THIS IS A BINARY SEARCH FOR THE CORRECT EXTENT OF THE REACTION

   EXTINC=0.25*EXTMAX
   EXTENT=0.5*EXTMAX
   B=RX(3)
   A=RX(4)+B
   AA=A*A
   BB=A*B
   CC=B*B
   DO 78 I=1,10
   A=1.0/(1.0-EXTENT)
   IF(FOR.GT.0.01) GO TO 72
   AMTINT=AA*A*EXTENT-2.0*BB*ALOG(A)+CC*EXTENT
   GO TO 76
72 IF(ABS(1.0-FOR).LT.0.01) GO TO 74
   B=1.0-FOR*EXTENT
   C=1.0/(1.0-FOR)
   AMTINT=(FOR-A*EXTENT-FOR/B+2.0*C*FOR*ALOG(A*B))*AA*C*C
   AMTINT=AMTINT-2.0*BB*C*(FOR*EXTENT/B-C*ALOG(A*B))-CC*EXTENT/B
   AMTINT=-AMTINT
   GO TO 76
74 AMTINT=AA*(A*A*A-1.0)/3.0-BB*(A*A-1.0)+CC*(A-1.0)
76 CONTINUE
   IF(AMTINT.GT.RK) EXTENT=EXTENT-EXTINC
   IF(AMTINT.LT.RK) EXTENT=EXTENT+EXTINC
   EXTINC=EXTINC/2.0
78 CONTINUE
```

```
      DELTAX=RX(3)*EXTENT
      DELTAY=0.0
      GO TO 100
   80 CONTINUE


   IRXN = 2  REACTION BETWEEN PU (IV) AND U (IV)
      RK = RATE CONSTANT
      RCU = RATIO OF PU (IV) TO REDUCTANT
      EXTMAX = MAXIMUM EXTENT BASED ON PU (IV) AND A TEN SECOND HALF TIME

      IF(RX(3).LT.1.0E-10.OR.RX(5).LT.1.0E-10) GO TO 90
      EXTMAX=AMIN1(1.0,1.0-0.5**(6.0*DTHETA),2.0*RX(5)/RX(3))
      RK=170.0
      RK=RK*DTHETA*RX(5)
      RCU=RX(3)/(2.0*RX(5))
      RHO=RX(1)*RX(1)
      REXP=1.0E20
      IF(RK*(1.0-RCU).LT.100.0*RHO) REXP=EXP(RK*(1.0-RCU)/RHO)
      EXTENT=RK/(RK+RHO)
      IF(ABS(1.0-RCU).GT.0.05) EXTENT=(1.0-REXP)/(RCU-REXP)
      EXTENT=AMIN1(EXTENT,EXTMAX)
      DELTAX=RX(3)*EXTENT
      DELTAY=0.0
      GO TO 100
   90 CONTINUE


   IRXN = 1  INSTANTANEOUS REDUCTION OF PU (IV)
      THIS MECHANISM IS USED BY ALL REDUCTION REACTIONS WHEN THE
      CONCENTRATIONS FALL BELOW A THRESHHOLD LEVEL

      DELTAX=AMIN1(-RX(3)/STOICX(3,IRXN),-RX(5)/STOICX(5,IRXN))
      DELTAY=0.0
  100 CONTINUE
      DO 105 I=1,6
      XS(J,KZ,I,2)=XS(J,KZ,I,2)+STOICX(I,IRXN)*DELTAX
      YS(J,KZ,I,2)=YS(J,KZ,I,2)+STOICY(I,IRXN)*DELTAY
      IF(XS(J,KZ,I,2).LE.1.E-20) XS(J,KZ,I,2)=0.0
      IF(YS(J,KZ,I,2).LE.1.E-20) YS(J,KZ,I,2)=0.0
      IF(XS(J,KZ,I,2).NE.0.0) ISOL(I)=1
      IF(YS(J,KZ,I,2).NE.0.0) ISOL(I)=1
      IF(ISOL(I).EQ.1.AND.NSOLU.LT.I) NSOLU=I
  105 CONTINUE
  150 CONTINUE
  300 RETURN
      END
      SUBROUTINE STARTS


   SUBROUTINE STARTS DOES GENERAL INPUT FUNCTIONS.  IT READS ALL THE
   INFORMATION REQUIRED FOR A TIME PERIOD, CONVERTS THE UNITS TO A
   MOLAL BASIS, AND SETS THE HYDRAULICS FOR THE SYSTEM.

      COMMON/CONTRL/ NTOST,NSOLU,CTBP,NEWIN,NEWOUT,SPH,IPNCH,IFAST,IRXN,
     .  DTHETA,IVOLM,IVOLS,IPRO,TEMPI,ISOL(6),NSTR,ISTR,JSTR,CODUM(100),
     .  IPROCE
      COMMON/CONCS/ X(6,100,2),Y(6,100,2),XS(100,3,6,2),YS(100,3,6,2)
      COMMON/DISTRB/ TEMPC,ARY(6),DTRY(6)
      COMMON/FLOWS/ A(100),O(100),AT(100),OT(100)
      COMMON/MOLALC/ AQ(6),OR(6),TEMP,CONVA,CONVO,TCONC
      COMMON/STREAM/ XFD(6,100),YFD(6,100),AFDRT(100),OFDRT(100),
     .   AFDTEM(100),OFDTEM(100),ALVRT(100),OLVRT(100)
```

```
      COMMON/TEMPS/ TPROF(100,2),ATS(100,3,2),OTS(100,3,2)
      COMMON/VOLS/ AVOL(100),OVOL(100),ASVOL(100),OSVOL(100)
      COMMON/ELECT/ XP(6,100),XE(6,99),DX(6,99),FR,DIV,DT,
     .              PS,QS,MS,VC,BRU,BRPU,OC,OA,GAM
      COMMON/RXNS/RXNTRM(6,100),AIN(6),OIN(6)
      DIMENSION CON(6)
      INTEGER PS,QS,DIV


EQCKDG IS A CHECK ON THE INITIAL PROFILE.  IF EQCKDG = 1.0 THE PROFILE
WAS CREATED BY THE PROGRAM.  OTHERWISE, THE PROFILE CONCENTRATIONS
ARE ADJUSTED SO THAT THE PHASES ARE IN EQUILIBRIUM.

      EQCKDG=0.0


TCONC IS AN INDICATOR FOR THE MOLAL CONVERSION.  TCONC = -1 SINCE
MOLAR CONCENTRATIONS WILL BE PASSED TO THE MOLAL SUBROUTINE.

      TCONC=-1.0


      DO 5 I=1,6
      AQ(I)=0.0
      OR(I)=0.0
    5 CONTINUE


NEWIN DETERMINES IF NEW INPUT STREAMS ARE TO BE SPECIFIED.  NEWIN = 0
INDICATES THAT NEW INPUT FLOWS WILL NOT BE GIVEN SO THE APPROPRIATE
SECTION OF THE PROGRAM SHOULD BE BYPASSED.

      IF(NEWIN.EQ.0) GO TO 40

THIS SECTION RE-INITIALIZES AND INPUTS ALL THE FEED STREAM INFORMATION.


      DO 10  J=1,NTOST
      AFDRT(J)=0.0
      OFDRT(J)=0.0
      AFDTEM(J)=TEMPI
      OFDTEM(J)=TEMPI
      DO 10 I=1,NSOLU
      XFD(I,J)=0.0
      YFD(I,J)=0.0
   10 CONTINUE
   15 CONTINUE
      READ (21,1001) J,JHAS,FDRT,(CON(I),I=1,6),TEMP,INDEX
      IF(J.LE.0) J=100
      IF(JHAS.EQ.1) GO TO 20
      OFDRT(J)=FDRT
      IF(TEMP.NE.0.) OFDTEM(J)=TEMP
      DO 17 I=1,6
      IF(CON(I).NE.0.0) ISOL(I)=1
      YFD(I,J)=CON(I)
   17 CONTINUE
      GO TO 30
   20 CONTINUE
      AFDRT(J)=FDRT
      IF(TEMP.NE.0.) AFDTEM(J)=TEMP
      IF(IRXN.GE.4) ISOL(2)=1
      IF(IRXN.GE.4) ISOL(5)=1
      DO 25 I=1,6
```

```
   IF(CON(I).NE.0.0) ISOL(I)=1
   XFD(I,J)=CON(I)
25 CONTINUE
30 CONTINUE
   IF(INDEX.EQ.1) GO TO 15
40 CONTINUE
```

NEWOUT DETERMINES IF NEW PRODUCT STREAMS ARE TO BE SPECIFIED. NEWOUT = 0 INDICATES THAT NO NEW PRODUCT STREAMS WILL BE SPECIFIED, SO THIS SECTION OF THE PROGRAM WILL BE BYPASSED.

```
   IF(NEWOUT.EQ.0) GO TO 60
```

THIS SECTION RE-INITIALIZES AND INPUTS ALL THE PRODUCT STREAM INFORMATION.

```
   DO 45 J=1,NTOST
   ALVRT(J)=0.0
   OLVRT(J)=0.0
45 CONTINUE
50 CONTINUE
   READ (21,2001) J,JHAS,OTRT,INDEX
   IF(J.LE.0) J=100
   IF(JHAS.EQ.1) ALVRT(J)=OTRT
   IF(JHAS.EQ.0) OLVRT(J)=OTRT
   IF(INDEX.EQ.1) GO TO 50
60 CONTINUE
```

IPRO DETERMINES IF AN INITIAL PROFILE IS TO BE READ OFF CARDS. IF NOT (IPRO = 0) THIS SECTION OF THE PROGRAM IS BYPASSED.

```
   IF(IPRO.EQ.0) GO TO 80
```

THIS SECTION RE-INITIALIZES THE CONCENTRATION PROFILES WITH THE VALUES READ OFF OF THE PROFILE CARDS. SETTLER CONCENTRATIONS ARE ASSUMED TO BE THE SAME AS THE MIXER CONCENTRATIONS FOR A STAGE.

```
   DO 70 J=1,NTOST
   READ(21,1002) (X(I,J,2),I=1,6),TPROF(J,2)
   READ(21,1002) (Y(I,J,2),I=1,6),EQCKDG
   IF(TPROF(J,2).EQ.0.0) TPROF(J,2)=TEMPI
   TPROF(J,1)=TPROF(J,2)
   DO 65 I=1,6
   RXNTRM(I,J)=0.0
   IF(X(I,J,2).NE.0.0) ISOL(I)=1
   IF(Y(I,J,2).NE.0.0) ISOL(I)=1
65 CONTINUE
   DO 70 K=1,3
   DO 70 L=1,2
   ATS(J,K,L)=TPROF(J,1)
   OTS(J,K,L)=TPROF(J,1)
70 CONTINUE
80 CONTINUE
```

NSOLU IS SET TO THE SUBSCRIPT OF THE HIGHEST NUMBERED COMPONENT IN THE SYSTEM. ISOL INDICATES WHETHER A COMPONENT IS PRESENT. THIS CHECK SAVES TIME BY BYPASSING CALCULATIONS WHICH WOULD ALWAYS RESULT IN ZERO CONCENTRATION. THUS IF ONLY NITRIC ACID IS PRESENT, CALCULATIONS FOR THE NON-EXISTANT PLUTONIUM CAN BE SKIPPED IN SOME PLACES.

```
      NSOLU=1
      DO 82 I=1,6
      IF(ISOL(I).EQ.1) NSOLU=I
   82 CONTINUE


   CONVRT PRINTS THE FEED STREAM INFORMATION, AND CONVERTS THE CONCENTRATIONS
   TO MOLAL UNITS.

      IF(NEWIN+IPRO.EQ.0) GO TO 120
      CALL CONVPU
  120 CONTINUE



      IF(NEWOUT.EQ.0) GO TO 122


   PRINTING THE PRODUCT STREAM FLOWRATES.  THE PRINTED RATES MAY NOT BE
   THE VALUES USED IN CALCULATIONS DUE TO THE REQUIREMENT THAT NO MORE
   OF A PHASE MAY FLOW OUT OF A STAGE THAN FLOWED IN.

      DO 121 J=1,NTOST
      IF(ALVRT(J).NE.0.0) WRITE (6,1007)J,ALVRT(J)
      IF(OLVRT(J).NE.0.0) WRITE (6,1008)J,OLVRT(J)
  121 CONTINUE
  122 CONTINUE
      IF(NEWIN+NEWOUT.EQ.0) GO TO 127

   SET AQUEOUS AND ORGANIC INTERSTAGE FLOW (A AND O)

   A IS AQUEOUS INTERSTAGE FLOW
   O IS ORGANIC INTERSTAGE FLOW
   AT IS TOTAL AQUEOUS FLOW WITHIN A STAGE
   OT IS TOTAL ORGANIC FLOW WITHIN A STAGE


      IF(ALVRT(1).GT.AFDRT(1)) ALVRT(1)=AFDRT(1)
      A(1)=AFDRT(1)-ALVRT(1)
      AT(1)=AFDRT(1)
      FR=A(1)
      IF(OLVRT(NTOST).GT.OFDRT(NTOST)) OLVRT(NTOST)=OFDRT(NTOST)
      O(NTOST)=OFDRT(NTOST)-OLVRT(NTOST)
      OT(NTOST)=OFDRT(NTOST)
      DO 125 I=2,NTOST
      IF(ALVRT(I).GE.A(I-1)+AFDRT(I)) ALVRT(I)=A(I-1)+AFDRT(I)
      A(I)=A(I-1)+AFDRT(I)-ALVRT(I)
      AT(I)=A(I)+ALVRT(I)
      NST = NTOST+1-I
      IF(OLVRT(NST).GE.O(NST+1)+OFDRT(NST)) OLVRT(NST)=O(NST+1)+
     .     OFDRT(NST)
      O(NST) = O(NST+1)+OFDRT(NST)-OLVRT(NST)
      IF(NSTR.EQ.1.AND.NST.EQ.ISTR-1) O(NST)=OFDRT(NST)-OLVRT(NST)
      IF(NSTR.EQ.1.AND.NST.EQ.JSTR) O(NST)=O(ISTR)+OFDRT(NST)-OLVRT(NST)
     .  +O(NST+1)
      OT(NST)=O(NST)+OLVRT(NST)

   THE INTERSTAGE FLOWS ARE NOW SET

  125 CONTINUE
  127 CONTINUE
```

```
 IVOLM AND IVOLS ARE NOW CHECKED TO SEE IF ANY VOLUME CARDS ARE TO BE
 READ, OR IF THE VOLUMES ARE EVEN GOING TO BE CHANGED

    IF(IVOLS+IVOLM.EQ.0) GO TO 310
    DO 128 J=1,NTOST
    IF(IVOLM.NE.0) AVOL(J)=0.0
    IF(IVOLM.NE.0) OVOL(J)=0.0
    IF(IVOLS.NE.0) ASVOL(J)=0.0
    IF(IVOLS.NE.0) OSVOL(J)=0.0
128 CONTINUE
    IF(IVOLM.EQ.3.AND.IVOLS.EQ.3) GO TO 200
    IF(IVOLM.EQ.3.AND.IVOLS.EQ.0) GO TO 200
    IF(IVOLM.EQ.0.AND.IVOLS.EQ.3) GO TO 200

 VOLUMES ARE NOW READ IN.  TO MINIMIZE THE INPUT CARDS NEEDED, THE VOLUMES
       NEED BE SPECIFIED ONLY WHEN THE VOLUME CHANGES FROM THAT OF THE
       PREVIOUS STAGE.

130 READ(21,2101) ISEC,I,VOLA,VOLO,INDX
    IF(I.LE.0) I=100
    IF(ISEC.EQ.1.AND.IVOLM.EQ.0) GO TO 145
    IF(ISEC.EQ.2.AND.IVOLS.EQ.0) GO TO 145
    IF(ISEC.EQ.2) GO TO 140
    AVOL(I)=VOLA
    OVOL(I)=VOLO
    GO TO 145
140 ASVOL(I)=VOLA
    OSVOL(I)=VOLO
145 IF(INDX.EQ.1) GO TO 130


 THE VOLUMES ARE NOW PROPAGATED THROUGH OUT THE COLUMN IN THE MANNER CHOSEN
 ALL VOLUMES AND FLOWS ARE PRINTED IF ANY CHANGES WERE MADE

200 IF(AVOL(1).EQ.0.0) AVOL(1)=AT(1)
    IF(OVOL(1).EQ.0.0) OVOL(1)=OT(1)
    IF(ASVOL(1).EQ.0.0) ASVOL(1)=AT(1)
    IF(OSVOL(1).EQ.0.0) OSVOL(1)=OT(1)
    DO 300 I=1,NTOST
    J=I-1
    IF(IVOLM.EQ.0) GO TO 240
    GO TO (210,220,230),IVOLM
210 IF(AVOL(I).EQ.0.0) AVOL(I)=AVOL(J)
    IF(OVOL(I).EQ.0.0) OVOL(I)=OVOL(J)
    GO TO 240
220 IF(AVOL(I).EQ.0.0) AVOL(I)=AVOL(J)+OVOL(J)
    OVOL(I)=AVOL(I)*OT(I)/(AT(I)+OT(I))
    AVOL(I)=AVOL(I)-OVOL(I)
    GO TO 240
230 AVOL(I)=AT(I)
    OVOL(I)=OT(I)
240 IF(IVOLS.EQ.0) GO TO 280
    GO TO (250,260,270),IVOLS
250 IF(ASVOL(I).EQ.0.0) ASVOL(I)=ASVOL(J)
    IF(OSVOL(I).EQ.0.0) OSVOL(I)=OSVOL(J)
    GO TO 280
260 IF(ASVOL(I).EQ.0.0) ASVOL(I)=ASVOL(J)+OSVOL(J)
    OSVOL(I)=ASVOL(I)*OT(I)/(AT(I)+OT(I))
    ASVOL(I)=ASVOL(I)-OSVOL(I)
    GO TO 280
270 ASVOL(I)=AT(I)
    OSVOL(I)=OT(I)
```

```
280 IF(AVOL(I).LT.1.0E-4) AVOL(I)=0.0
    IF(OVOL(I).LT.1.0E-4) OVOL(I)=0.0
    IF(ASVOL(I).LT.1.0E-4) ASVOL(I)=0.0
    IF(OSVOL(I).LT.1.0E-4) OSVOL(I)=0.0
300 CONTINUE
310 IF(NEWIN+NEWOUT+IVOLM+IVOLS.EQ.0) GO TO 400
    WRITE (6,1106)
    WRITE (6,1107)
    WRITE (6,1108)
    DO 320 I=1,NTOST

 ADDING VOLUME OF ELECTROCELL,HEREBY THE ELECTROCELL IS TREATED
 AS A PIPE CONNECTED BETWEEN TWO CONSECUTIVE STAGES

    IF((I.EQ.PS).OR.(I.EQ.QS)) ASVOL(I)=ASVOL(I)+VC

    WRITE (6,1101)I,AVOL(I),OVOL(I),ASVOL(I),OSVOL(I),AT(I),OT(I),
   . A(I),O(I)
320 CONTINUE


    THIS SECTION INSURES THAT THE PHASES ARE IN CHEMICAL EQUILIBRIUM
    BEFORE THE ITERATIONS START.  THE CHECK IS DISABLED WHEN THE INITIAL
    PROFILE WAS CREATED BY THE PROGRAM, OR WHEN THE PROFILE WAS FROM
    CARDS PUNCHED BY THE PROGRAM.


    IF(IPRO.EQ.0.OR.EQCKDG.EQ.1.0) GO TO 400
    DO 350 J=1,NTOST
    AV=AVOL(J)+ASVOL(J)
    OV=OVOL(J)+OSVOL(J)
    DO 340 K=1,3
    DO 330 I=1,6
330 ARY(I)=X(I,J,2)
    ICK=0
    TEMPC=TPROF(J,2)
    CALL UCORPU
    DO 335 I=1,NSOLU
    IF(ISOL(I).EQ.0) GO TO 335
    X(I,J,1)=(OV*Y(I,J,2)+AV*X(I,J,2))/(AV+OV*DTRY(I))
    Y(I,J,1)=Y(I,J,2)+(X(I,J,2)-X(I,J,1))*AV/OV
    IF(ABS(X(I,J,1)-X(I,J,2)).GT.0.01*X(I,J,1)) ICK=1
    X(I,J,2)=X(I,J,1)
    Y(I,J,2)=Y(I,J,1)
335 CONTINUE
    IF(ICK.EQ.0) GO TO 345
340 CONTINUE
345 DO 350 K=1,3
    DO 350 L=1,2
    ATS(J,K,L)=TPROF(J,1)
    OTS(J,K,L)=TPROF(J,1)
    DO 350 I=1,NSOLU
    XS(J,K,I,L)=X(I,J,1)
    YS(J,K,I,L)=Y(I,J,1)
350 CONTINUE
400 RETURN


001 FORMAT(2I2,8F8.0,I2)
002 FORMAT(8F10.0)
007 FORMAT(14X,'AQUEOUS ',I3,5X,'PRODUCT STREAM REMOVED (FLOW RATE IN
   .MOLAL UNITS)',34X,1PE10.3)
008 FORMAT(14X,'ORGANIC ',I3,5X,'PRODUCT STREAM REMOVED (FLOW RATE IN
```

```
     .MOLAL UNITS)',34X,1PE10.3)
1101 FORMAT(8X,I3,2X,8('^',2X,1PE10.3,2X))
1106 FORMAT(///7X,'STAGE ^',4X,'MIXER VOLUME BY PHASE',4X,'^   SETTLER VO
     .VOLUME BY PHASE   ^',7X,'MIXER FLOW RATE',7X,'^',4X,'INTERSTAGE FL
     .OW RATE')
1107 FORMAT(8X,'NO.  ',4('^   AQUEOUS     ^   ORGANIC     '))
1108 FORMAT(13X,8('^',14X))
2001 FORMAT(2I2,F8.0,I2)
2101 FORMAT(2I2,2F8.0,I2)
     END
     SUBROUTINE PRTPU


  PRTOUT CONVERTS THE CONCENTRATIONS FROM THE MOLAL FORM USED BY THE PROGRAM
  TO THE MORE COMMON UNITS IN THE OUTPUT.  IN ORDER TO GET THE PROPER
  HEADINGS FOR THE PUREX SYSTEM, THE SUBPROGRAM ALSO PRINTS THE PROFILES.

     COMMON/CONTRL/ NTOST,NSOLU,CTBP,NEWIN,NEWOUT,SPH,IPNCH,IFAST,IRXN,
    .  DTHETA,IVOLM,IVOLS,IPRO,TEMPI,ISOL(6),NSTR,ISTR,JSTR,CODUM(100),
    .   IPROCE
     COMMON/CONCS/ X(6,100,2),Y(6,100,2),XS(100,3,6,2),YS(100,3,6,2)
     COMMON/FLOWS/ A(100),O(100),AT(100),OT(100)
     COMMON/MOLALC/ AQ(6),OR(6),TEMP,CONVA,CONVO,TCONC
     COMMON/STREAM/ XFD(6,100),YFD(6,100),AFDRT(100),OFDRT(100),
    .   AFDTEM(100),OFDTEM(100),ALVRT(100),OLVRT(100)
     COMMON/TEMPS/ TPROF(100,2),ATS(100,3,2),OTS(100,3,2)
     COMMON/ELECT/ XP(6,100),XE(6,99),DX(6,99),FR,DIV,DT,
    .              PS,QS,MS,VC,BRU,BRPU,OC,OA,GAM
     DIMENSION AOUT(100),OOUT(100)
     INTEGER PS,QS,DIV
     TCONC=1.0
     WRITE (6,1101)
     WRITE (6,1102)
     WRITE (6,1103)
     WRITE (6,1104)
     DO 20 J=1,NTOST
     DO 10 I=1,NSOLU
     AQ(I)=X(I,J,2)
     OR(I)=Y(I,J,2)
 10  CONTINUE
     TEMP=TPROF(J,2)
     CALL MOLALP
     ADEN=(1000.-72.4*AQ(2)-130.*(AQ(3)+AQ(4))-30.9*AQ(1)-31.*AQ(6))/
    .     (1000./0.99707)+0.39404*AQ(2)+0.49202*(AQ(3)+AQ(4))
    .     +0.06301*AQ(1)+0.213*AQ(6)
     AOUT(J)=AT(J)*CONVA
     OOUT(J)=OT(J)*CONVO
     DO 15 I=1,NSOLU
     X(I,J,2)=AQ(I)/CONVA
     Y(I,J,2)=OR(I)/CONVO
 15  CONTINUE
     X(2,J,2)=X(2,J,2)*238.
     Y(2,J,2)=Y(2,J,2)*238.
     X(3,J,2)=X(3,J,2)*239.
     Y(3,J,2)=Y(3,J,2)*239.
     X(4,J,2)=X(4,J,2)*239.
     Y(4,J,2)=Y(4,J,2)*239.
     X(5,J,2)=X(5,J,2)*238.
     Y(5,J,2)=Y(5,J,2)*238.
     WRITE (6,1000)J,(X(I,J,2),I=1,6),ADEN,AOUT(J),TPROF(J,2)

  PRINTING THE CONCENTRATIONS LEAVING THE ELECTROCELL. NOTE THAT
  THE UNITS USED IN THE ELECTROCELL ARE MOLAR,SO
```

```
NO CONVERSION IS NEEDED.

      IF((J.NE.PS).OR.(IRXN.LT.4)) GO TO 18
         XP(2,PS)=XP(2,PS)*238.
         XP(3,PS)=XP(3,PS)*239.
         XP(4,PS)=XP(4,PS)*239.
         XP(5,PS)=XP(5,PS)*238.
         WRITE(6,1002)J,(XP(I,PS),I=1,6)
   18 IF((J.NE.QS).OR.(IRXN.LT.4)) GO TO 20
         XP(2,QS)=XP(2,QS)*238.
         XP(3,QS)=XP(3,QS)*239.
         XP(4,QS)=XP(4,QS)*239.
         XP(5,QS)=XP(5,QS)*238.
         WRITE(6,1002)J,(XP(I,QS),I=1,6)

      IF(ALVRT(J).EQ.0.0) GO TO 20
         CONVA=CONVA*ALVRT(J)
         WRITE (6,1001)J,CONVA
   20 CONTINUE
      WRITE (6,1105)
      WRITE (6,1106)
      WRITE (6,1107)
      WRITE (6,1104)
      DO 30 J=1,NTOST
      ODEN=(1000.-17.4*0.29-97.*Y(2,J,1)-139.*(Y(3,J,1)+Y(4,J,1))
     .     -43.*Y(1,J,1))*(1.0+1.8)/((273.6/266.32+227.5/170.34*1.8)
     .     *1000.)+0.01802*.29+.39404*Y(2,J,1)+.49202*(Y(3,J,1)
     .     +Y(4,J,1))+.06301*Y(1,J,1)
      IF(OLVRT(J).EQ.0.0) GO TO 28
      CONVO=OLVRT(J)*OOUT(J)/OT(J)
      WRITE (6,1001)J,CONVO
   28 WRITE(6,1000)J,(Y(I,J,2),I=1,6),ODEN,OOUT(J),
     /  CODUM(J)
   30 CONTINUE

      DO 50 K=1,100

   50 CONTINUE

1000 FORMAT(12X,I3,1X,9('^ ',1PE10.3,1X))
1001 FORMAT(12X,I3,' ^ PRODUCT STREAM ',74X,'^ ',1PE10.3,' ^')
1002 FORMAT(11X,'E',I3,1X,6('^ ',1PE10.3,1X))
1101 FORMAT(5X,'AQUEOUS PHASE')
1102 FORMAT(10X,'STAGE ^ NITRIC ACID^   U (VI)   ^  PU (IV)   ^  PU (II
     .I)  ^   U (IV)   ^ NITRATE ION^  DENSITY   ^ MIXER FLOW ^TEMPERATU
     .RE')
1103 FORMAT(11X,'NO.  ^   (MOL/L)  ^   (G/L)    ^   (G/L)    ^   (G/L)
     .    ^   (G/L)    ^  (MOL/L)   ^   (G/ML)   ^  (L/MIN)   ^(CENTIGRA
     .DE)')
1104 FORMAT(16X,9('^',12X))
1105 FORMAT('     ORGANIC PHASE')
1106 FORMAT(10X,'STAGE ^ NITRIC ACID^   U (VI)   ^  PU (IV)   ^  PU (II
     .I)  ^   U (IV)   ^ NITRATE ION^  DENSITY   ^ MIXER FLOW ^ INVENTOR
     .Y')
1107 FORMAT(11X,'NO.  ^   (MOL/L)  ^   (G/L)    ^   (G/L)    ^   (G/L)
     .    ^   (G/L)    ^  (MOL/L)   ^   (G/ML)   ^  (L/MIN)   ^  CHANGE (
     .%)')
      RETURN
      END


      SUBROUTINE ELECTRO
```

```
      SUBROUTINE ELECTRO CALCULATES THE CONCENTRATION PROFILES FOR
      U(VI), U(IV), PU(IV), PU(III), HNO3, AND OTHER NITRATE SALTS
      IN AN ELECTROCELL


      DESCRIPTION OF VARIABLES IN ELECTRO

      XE(I,DIV)     CONCENTRATION OF COMPONENT I IN SUBDIVISION DIV
      XP(I,MS)      CONCENTRATION OF COMPONENT I LEAVING THE ELECTROCELL;
                    USED ONLY IN THE SUBROUTINE PRTPU
      FR            FLOW RATE
*     MS            ELECTROCELL CONNECTING STAGE
*     DIV           NUMBER OF CELL SUBDIVISIONS
*     VC            VOLUME OF ELECTROCELL
*     BRU           ELECTRICAL FACTOR FOR U(VI)
*     BRPU          ELECTRICAL FACTOR FOR PU(IV)
*     OC            GEOMETRIC FACTOR OF THE CATHODE
*     OA            GEOMETRIC FACTOR OF THE ANODE
*     GAM           REACTION RATE CONSTANT FOR THE U(IV)+PU(IV) REACTION

      ( * MEANS THOSE VARIABLES MUST BE SPECIFIED IN THE INPUT FILE )


      COMMON/CONCS/ X(6,100,2),Y(6,100,2),XS(100,3,6,2),YS(100,3,6,2)
      COMMON/FLOWS/ A(100),O(100),AT(100),OT(100)
      COMMON/MOLALC/ AQ(6),OR(6),TEMP,CONVA,CONVO,TCONC
      COMMON/STREAM/ XFD(6,100),YFD(6,100),AFDRT(100),OFDRT(100),
     .   AFDTEM(100),OFDTEM(100),ALVRT(100),OLVRT(100)
      COMMON/ELECT/ XP(6,100),XE(6,99),DX(6,99),FR,DIV,DT,
     .              PS,QS,MS,VC,BRU,BRPU,OC,OA,GAM
      INTEGER PS,QS,DIV

      UNIT CONVERSION FROM MOLAL TO MOLAR

      TCONC=1.0

      DO 12 I=1,6
         AQ(I)=XS(MS,3,I,2)
         OR(I)=YS(MS,3,I,2)
12    CONTINUE

      CALL MOLALP
      FR=AT(MS)*CONVA
      DT=VC/(FR*DIV)

      DO 15 I=1,6
         XE(I,1)=AQ(I)/CONVA
15    CONTINUE

      IF (XE(1,1).LT.1.0E-12) GO TO 220


      CALCULATE THE CONCENTRATION CHANGES DUE TO ELECTROLYTIC
      REDUCTION OF U(VI) AND PU(IV) AT THE CATHODE AND THE
      CHEMICAL REACTION BETWEEN U(IV) AND PU(IV).

      DO 100 K=2,DIV
      DX(2,K)=-XE(2,K-1)*(1-EXP(-BRU*OC*DT))+GAM*
     *XE(5,K-1)*XE(3,K-1)/XE(1,K-1)**2*DT
      XE(2,K)=XE(2,K-1)+DX(2,K)
      DX(5,K)=-DX(2,K)
```

```
      XE(5,K)=XE(5,K-1)+DX(5,K)
      DX(3,K)=-XE(3,K-1)*(1-EXP(-BRPU*OC*DT))-2*GAM*XE(5,K-1)*
     *XE(3,K-1)/XE(1,K-1)**2*DT
      XE(3,K)=XE(3,K-1)+DX(3,K)
      IF(XE(3,K).LT.0) XE(3,K)=0.0
      DX(4,K)=-DX(3,K)
      XE(4,K)=XE(4,K-1)+DX(4,K)
      DX(1,K)=2*DX(2,K)
      XE(1,K)=XE(1,K-1)+DX(1,K)
      XE(6,K)=XE(6,1)
100   CONTINUE

      UNIT CONVERSION FROM MOLAR TO MOLAL

      TCONC=-1.0

      DO 105 I=1,6
         AQ(I)=XE(I,DIV)
         OR(I)=0.0
105   CONTINUE

      CALL MOLALP

      DO 110 I=1,6
           XP(I,MS)=XE(I,DIV)
           XS(MS,3,I,1)=XE(I,DIV)*CONVA
110   CONTINUE


120   RETURN
      END
```

DESCRIPTION OF INPUT FILE VARIABLES

Line No.	Column No.	Description of Variables
Start of the next case:		
1		FORMAT (212, 2F8.0, 4I2)
	1- 2	NTTL = number of title lines (up to ten).
	3- 4	NTOST = number of stages (up to 100).
	5-12	CTBP = volume fraction of TBP in the organic phase.
	13-20	TEMPI = initial and default temperature.
	21-22	NSTR = special piping indicator. = 1 for special routing indicated by ISTR and JSTR. = 0 otherwise
	23-24	ISTR = stage number.
	25-26	JSTR = stage number.
		When special piping is used, the organic stream for stage ISTR is fed to stage JSTR.
	27-28	IRXN = indication of reaction rate equation being used. = 0 No reactions will be considered = 1 Instantaneous reduction of plutonium = 2 Reduction of plutonium by uranium (IV) = 3 Reduction of plutonium by hydroxylamine = 4 Electrocell without chemical reaction = 5 Electrocell plus reduction by hydroxylamine = 6 Electrocell plus reduction by U(IV) = 7 Electrocell plus instantaneous reduction

2 Format (I2, I2, I3, F8.3,5F9.6)

1-2	PS	=	location of first electrocell (the first cell is connected between stage PS and stage PS+1)
3-4	QS	=	location of second electrocell (the second cell is connected between stage QS and stage QS+1)
5-7	DIV	=	number of cell subdivisions, the maximum number allowed is 99
8-15	VC	=	volume of electrocell in liters
16-24	BRU	=	electrical factor for uranium
25-33	BRPU	=	electrical factor for plutonium
34-42	OC	=	geometric factor for cathode
43-51	OA	=	geometric factor for anode
52-60	GAM	=	chemical reaction rate constant for the reaction $U^4 + 2Pu^4 = U^6 + 2Pu^3$

3 FORMAT (10A8)

1-80	Case title to be printed. Up to ten lines which describe the case.

After the title lines have been red, the new case is begun. All volumes, flows, and concentrations are set to zero.

4 FORMAT (4F8.0, 7I2)

1-8	DTHETA	=	time increment (min).
9-16	DPRINT	=	time between successive printings of the concentration profile.
17-24	TSTOP	=	time when the calculations will stop and a new line No. 4 will be read to start a new time period or case.
25-32	TOL	=	tolerance (percent per minute). When the tolerance is met in every stage, a new line No. 4 will be read to start a new time period or case.

34 NEWIN = 1, if new feed streams are to be specified.
= 0, if the present feed streams are to be continued

36 NEWOUT = 1, if new product streams are to be specified.
= 0, if the present product streams are to be continued.

38 IVOLM specifies how mixer volumes will be given.

= 0, if the present mixer volumes will be continued.
= 1, if the new volumes are to be given by phase.
= 2, if the new volumes are to be given as a total volume, with the phase split according to the flow ratio.
= 3, if the new volumes are to be given by the phase flow multiplied by a unit time.

40 IVOLS specifies how the settler volumes will be given. The indicators are identical to those for IVOLM.

42 IPRO = 0, if the present concentration profile is to be continued.
= 1, if a new initial profile is to be read in.

44 IFAST = 0, if the Runge-Kutta integration is to be used.
= 1, if the faster, but less accurate, integration is desired.

46 IPNCH = 0, if printout is not wanted.
= 1, if concentration profile is to be printed when TSTOP or TOL is reached.

5 FORMAT (2I2, 8F8.0, I2)

Feed stream lines which will be read only if NEWIN = 1.

1-2	I	=	stage number where this feed will enter.
4	JHAS	=	0 for organic feed streams.
		=	1 for aqueous feed streams.
5-12	FDRT	=	flow rate (liters/min).
13-20	CON1	=	nitric acid concentration (M).
21-28	CON2	=	uranium concentration (g/liter).
29-36	CON3	=	plutonium (IV) concentration (g/liter).
37-44	CON4	=	plutonium (III) concentration (g/liter).
45-52	CON5	=	plutonium reductant concentration (M).
53-60	CON6	=	inextractable nitrate ion concentration (M)
61-68	TEMP	=	feed temperature (°C). If TEMP = 0.0, the default temperature (TEMPI) is used.
70	INDEX	=	0, if this is the last feed-stream line.
		=	1, if more feed-stream line follow.

6 FORMAT (2I2, F8.0, I2).

Product stream line which will be read only if NEWOUT = 1.

1-2	I	=	stage number where the stream will leave.
4	JHAS	=	0 for an organic product stream.
		=	1 for an aqueous product stream.
5-12	OTRT	=	flow rate of product stream. If OTRT is larger than the appropriate flow through stage I, the exiting rate will be set so that the entire phase, but no more, is removed.

14	INDEX	=	0, if no more product-stream line follow.
		=	1, if more product-stream line follow.

7 FORMAT (7F10.0)

Initial profile line which will be read only if IPRO = 1. An aqueous line No. 7 followed by an organic card No. 8 is required for every stage.

1-10	CON1	=	nitric acid concentration (M)
11-20	CON2	=	uranium concentration (g/liter).
21-30	CON3	=	plutonium (IV) concentration (g/liter).
31-40	CON4	=	plutonium (III) concentration (g/liter).
41-50	CON5	=	plutonium reductant concentration (M)
51-60	CON6	=	inextractable nitrate concentration (M).
61-70	TPROF	=	stage temperature (°C).

8 FORMAT (7F10.0)

Organic profile line.

1-10	CON1	=	nitric acid concentration (M)
11-20	CON2	=	uranium concentration (g/liter).
21-30	CON3	=	plutonium (IV) concentration (g/liter).
31-40	CON4	=	plutonium (III) concentration (M).
41-50	CON5	=	plutonium reductant concentration (M).
51-60	CON6	=	inextractable nitrate concentration (M).
61-70	EQCKDG	=	This variable is a check on how the profile printout was created. The user should not put anything in this field. When the profiles are printed by the program, a 1.0 is put here to signify that the phases are in approximate chemical equilibrium.

9	FORMAT (2I2, 2F8.0, I2).		
	Volumes for mixers and settlers. These cards are needed only if IVOLM or IVOLS equals 1 or 2.		
2	ISEC	=	1 for mixer volumes
		=	2 for settler volumes.
3- 4	I	=	stage number.
5-12	VOLA		
	If IVOLM = 1 and ISEC = 1, or if IVOLS = 1 and ISEC = 2, VOLA = aqueous volume. However, if IVOLM = 2 and ISEC = 1, or if IVOLS = 2 and ISEC = 2, VOLA = total volume.		
13-20	VOLO = organic volume if IVOLM = 1 and ISEC = 1, or IVOLS = 1 and ISEC = 2.		
22	INDX	=	0 for the last volume line.
		=	1, if more volume lines follow.

After the last volume line has been read in, the program begins calculating concentrations. When the elapsed time is equal to TSTOP or the tolerance is met, a new line No. 4 is read. If this line is merely giving new DTHETA, DPRINT, TSTOP, TOL, IFAST, or IPNCH values, the calculation continues as before but with the new values. If this line requests that new feed streams, product streams, concentration profiles, or volumes be used, the desired information will be read in and the calculation restarted for the new time period. At the start of a new time period, the elapsed time will be set to 0.0.

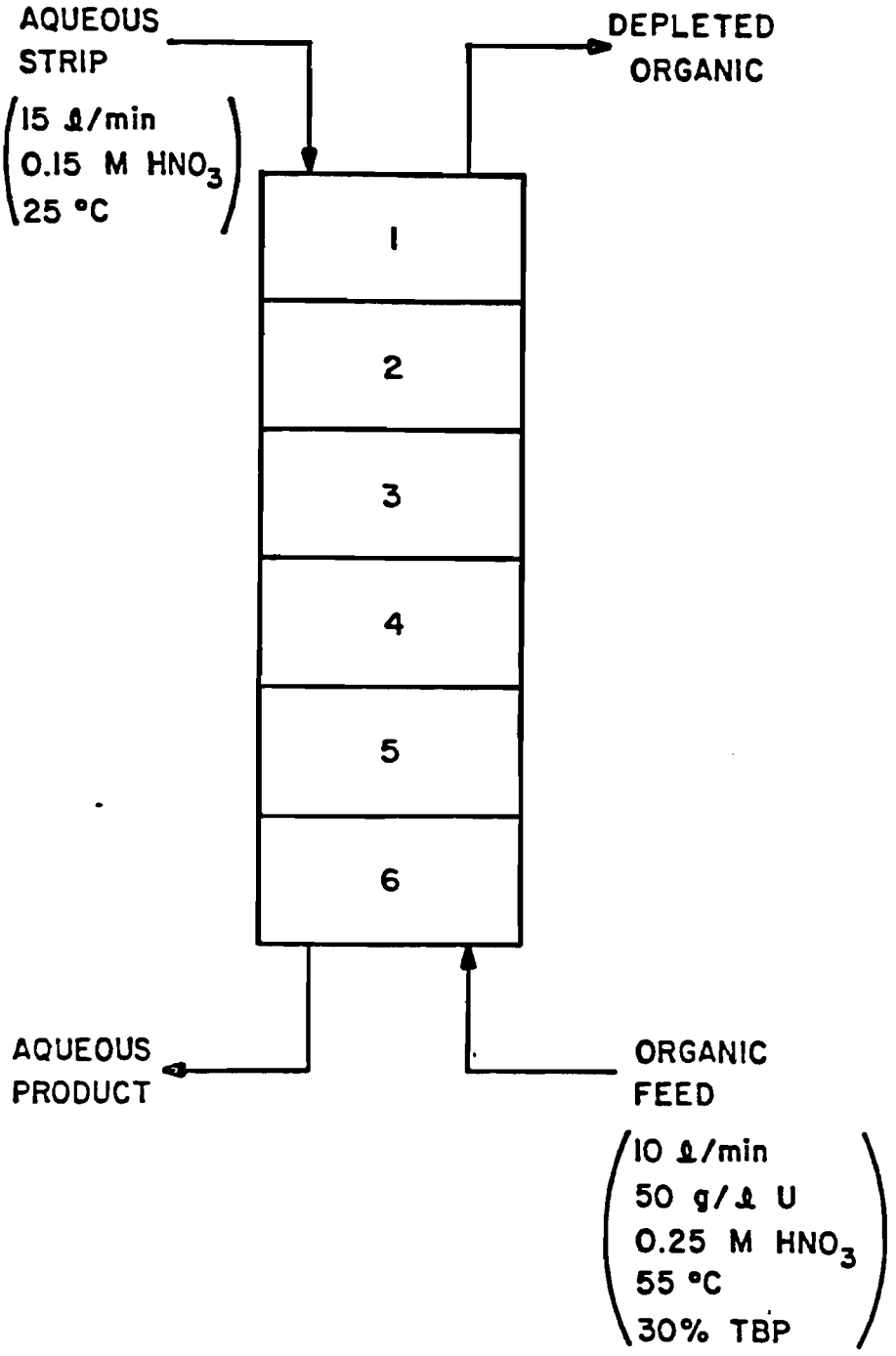

Figure C.1 Six-Stage Uranium Stripping Cascade

```
 TEST SAMPLE No. 1

INPUT FILE "SAMPLE1.INP"

 2 6 0.3    30.0     0 0 0 0
THIS IS A TEST EXAMPLE OF SEPHIS5-GT
CASE 1 - A SIX-STAGE URANIUM STRIPPING CASCADE
1.0      400.0    400.0 0.01     1 0 3 3 0 1 0
 1 1 15.0   0.15       0.0  0.0     0.0     0.0     0.0     25.0     1
 6 0 10.0   0.25      50.0  0.0     0.0     0.0     0.0     55.0     0
 0.0    0.0

 OUTPUT FILE "SAMPLE1.OUT"

 CALCULATIONS FOR A SOLVENT EXTRACTION PROCESS HAVING INTERACTING SOLUTES

 PUREX PROCESS
 THIS IS A TEST EXAMPLE OF SEPHIS5-GT
 CASE 1 - A SIX-STAGE URANIUM STRIPPING CASCADE
-DTHETA =    1.000 MINUTES PER TIME INCREMENT
 DPRINT =  400.000 MINUTES BETWEEN PRINTING OF PROFILES
 IFAST = 1  THE FAST INTEGRATION TECHNIQUE WILL BE USED
 THIS TIME PERIOD WILL END WHEN TIME = TSTOP =  400.000 MINUTES, OR A TOLERANCE OF TOL =   .0100 % PER MINUTE IS REACHED
 NUMBER OF STAGES =   6
 NEWIN = 1 NEW INPUT FLOWS WILL BE GIVEN
 NEWOUT = 0  OUTPUT STREAMS WILL BE UNCHANGED
 IVOLM = 3 MIXER VOLUMES DETERMINED BY PHASE FLOW
 IVOLS = 3 SETTLER VOLUMES GIVEN BY PHASE FLOW
 IPRO = 0  A NEW INITIAL PROFILE WILL NOT BE READ
 IPNCH = 0   NO PUNCHED CARD OUTPUT
 NSTR = 0   NO UNUSUAL ROUTING PATTERN
 TEMPI =  3.000E+01  INITIAL & DEFAULT TEMPERATURE
 IRXN = 0  NO REACTIONS WILL BE CONSIDERED
-FEED & PRODUCT      STAGE  NITRIC ACID     U (VI)        PU (IV)       PU (III)       U (IV)     NITRATE ION    FLOW RAT     TEMP
 STREAM DATA          NO.     (MOL/L)       (G/L)         (G/L)         (G/L)         (G/L)        (MOL/L)       (L/MIN)       (C)
           AQUEOUS    1     1.500E-01      .000E+00      .000E+00      .000E+00      .000E+00      .000E+00     1.500E+01      25.0
         30.0 % TBP   6     2.500E-01     5.000E+01      .000E+00      .000E+00      .000E+00      .000E+00     1.000E+01      55.0
```

STAGE NO.	MIXER VOLUME BY PHASE		SETTLER VOLUME BY PHASE		MIXER FLOW RATE		INTERSTAGE FLOW RATE	
	AQUEOUS	ORGANIC	AQUEOUS	ORGANIC	AQUEOUS	ORGANIC	AQUEOUS	ORGANIC
1	1.493E+01	9.655E+00	1.493E+01	9.655E+00	1.493E+01	9.655E+00	1.493E+01	9.655E+00
2	1.493E+01	9.655E+00	1.493E+01	9.655E+00	1.493E+01	9.655E+00	1.493E+01	9.655E+00
3	1.493E+01	9.655E+00	1.493E+01	9.655E+00	1.493E+01	9.655E+00	1.493E+01	9.655E+00
4	1.493E+01	9.655E+00	1.493E+01	9.655E+00	1.493E+01	9.655E+00	1.493E+01	9.655E+00
5	1.493E+01	9.655E+00	1.493E+01	9.655E+00	1.493E+01	9.655E+00	1.493E+01	9.655E+00
6	1.493E+01	9.655E+00	1.493E+01	9.655E+00	1.493E+01	9.655E+00	1.493E+01	9.655E+00

TRANSIENT BEHAVIOR RESULTS MIXER CONCENTRATION GIVEN

- TIME = .00 MINUTES

AQUEOUS PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	TEMPERATURE (CENTIGRADE)
1	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	1.493E+01	3.000E+01
2	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	1.493E+01	3.000E+01
3	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	1.493E+01	3.000E+01
4	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	1.493E+01	3.000E+01
5	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	1.493E+01	3.000E+01
6	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	1.493E+01	3.000E+01

ORGANIC PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	INVENTORY CHANGE %)
1	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	9.738E+00	2.000E+02
2	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	9.738E+00	2.000E+02
3	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	9.738E+00	2.000E+02
4	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	9.738E+00	2.000E+02
5	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	9.738E+00	2.000E+02
6	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	9.738E+00	2.000E+02

- TIME = 187.00 MINUTES

AQUEOUS PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	TEMPERATURE (CENTIGRADE)
1	1.508E-01	2.729E+00	.000E+00	.000E+00	.000E+00	.000E+00	1.006E+00	1.501E+01	2.500E+01
2	1.513E-01	7.563E+00	.000E+00	.000E+00	.000E+00	.000E+00	1.012E+00	1.504E+01	2.502E+01
3	1.513E-01	1.382E+01	.000E+00	.000E+00	.000E+00	.000E+00	1.021E+00	1.506E+01	2.507E+01
4	1.515E-01	2.077E+01	.000E+00	.000E+00	.000E+00	.000E+00	1.030E+00	1.510E+01	2.533E+01
5	1.607E-01	2.820E+01	.000E+00	.000E+00	.000E+00	.000E+00	1.041E+00	1.514E+01	2.649E+01
6	3.053E-01	3.181E+01	.000E+00	.000E+00	.000E+00	.000E+00	1.051E+00	1.522E+01	3.168E+01

OGANIC PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	INVENTORY CHANGE %)
1	1.061E-02	1.602E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.196E-01	9.749E+00	9.971E-03
2	1.202E-02	5.798E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.252E-01	9.763E+00	9.157E-03
3	1.306E-02	1.322E+01	.000E+00	.000E+00	.000E+00	.000E+00	8.352E-01	9.788E+00	6.673E-03
4	1.348E-02	2.279E+01	.000E+00	.000E+00	.000E+00	.000E+00	8.482E-01	9.821E+00	4.308E-03
5	1.428E-02	3.341E+01	.000E+00	.000E+00	.000E+00	.000E+00	8.626E-01	9.857E+00	2.515E-03
6	2.879E-02	4.471E+01	.000E+00	.000E+00	.000E+00	.000E+00	8.786E-01	9.900E+00	1.339E-03

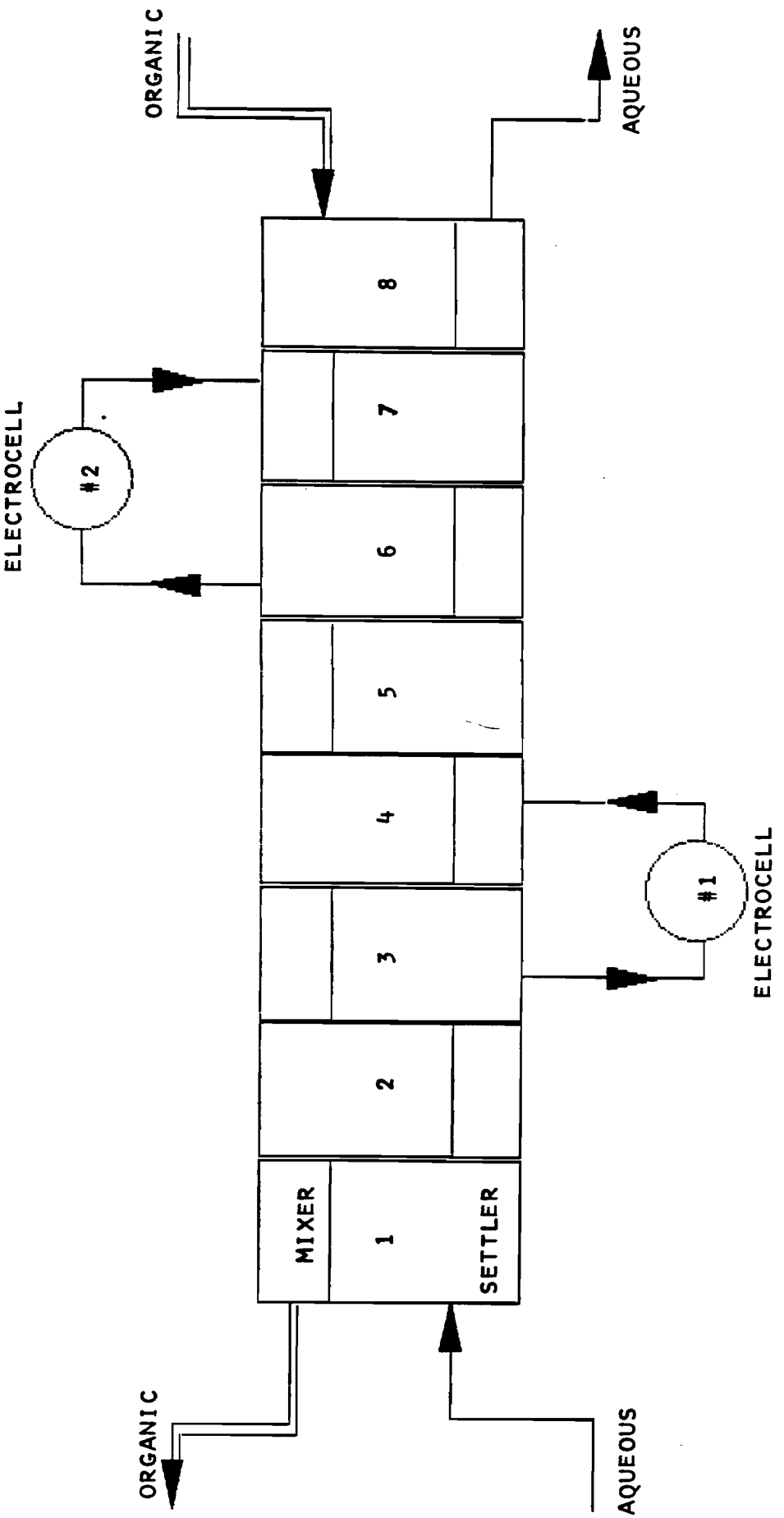

Figure C.2 Eight-Stage U-Pu Partitioning Cascade

```
EST SAMPLE No. 2

NPUT FILE "SAMPLE2.INP"

2 8 0.3    25.0     0 0 0 6
3 6 99   0.047 0.075   0.075    0.50     0.00     0.50
HIS IS A TEST EXAMPLE OF SEPHIS5-GT
ASE 2 - EIGHT-STAGE CONTACTOR WITH TWO ELECTROCELLS
.0      400.0    400.0 0.1      1 0 2 2 0 1 0
1 1  0.006 0.5      000.0  0.00    0.0     0.0     0.5     25.0     1
3 0  0.018 0.15      58.4  5.84    0.0     0.0     0.0     25.0     0
1 1 0.01            1
2 1 0.06            0
3.0    0.0

JTPUT FILE "SAMPLE2.OUT"

ALCULATIONS FOR A SOLVENT EXTRACTION PROCESS HAVING INTERACTING SOLUTES
PUREX PROCESS
THIS IS A TEST EXAMPLE OF SEPHIS5-GT
CASE 2 - EIGHT-STAGE CONTACTOR WITH TWO ELECTROCELLS
DTHETA =    1.000 MINUTES PER TIME INCREMENT
DPRINT =  400.000 MINUTES BETWEEN PRINTING OF PROFILES
IFAST = 1  THE FAST INTEGRATION TECHNIQUE WILL BE USED
THIS TIME PERIOD WILL END WHEN TIME = TSTOP =  400.000 MINUTES, OR A TOLERANCE OF TOL =   .1000 % PER MINUTE IS REACHED
NUMBER OF STAGES =   8
IEWIN = 1 NEW INPUT FLOWS WILL BE GIVEN
IEWOUT = 0  OUTPUT STREAMS WILL BE UNCHANGED
IVOLM = 2 TOTAL MIXER VOLUME GIVEN
IVOLS = 2 TOTAL SETTLER VOLUME GIVEN
IPRO = 0  A NEW INITIAL PROFILE WILL NOT BE READ
IPNCH = 0   NO PUNCHED CARD OUTPUT
ISTR = 0   NO UNUSUAL ROUTING PATTERN
TEMPI =  2.500E+01  INITIAL & DEFAULT TEMPERATURE
IRXN = 6  ELECTROCELL PLUS REDUCTION BY U(IV)
ELECTROCELL BETWEEN STAGE  3 AND STAGE  4
ELECTROCELL BETWEEN STAGE  6 AND STAGE  7
VC   =   .0470 LITER -- VOLUME OF ELECTROCELL
VRU  =   .0750       -- ELECTRICAL FACTOR FOR U(VI)
VRPU =   .0750       -- ELECTRICAL FACTOR FOR PU(IV)
XC   =   .5000       -- GEOMETRIC FACTOR OF CATHODE
XA   =   .0000       -- GEOMETRIC FACTOR OF ANODE
AM   =   .5000       -- REACTION RATE CONSTANT FOR U(IV)+ PU(IV) REACTION
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FEED & PRODUCT STREAM DATA	STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	FLOW RAT (L/MIN)	TEMP (C)
AQUEOUS	1	5.000E-01	.000E+00	.000E+00	.000E+00	.000E+00	5.000E-01	6.000E-03	25.0
30.0 % TBP	8	1.500E-01	5.840E+01	5.840E+00	.000E+00	.000E+00	.000E+00	1.800E-02	25.0

STAGE NO.	MIXER VOLUME BY PHASE		SETTLER VOLUME BY PHASE		MIXER FLOW RATE		INTERSTAGE FLOW RATE	
	AQUEOUS	ORGANIC	AQUEOUS	ORGANIC	AQUEOUS	ORGANIC	AQUEOUS	ORGANIC
1	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
2	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
3	2.512E-03	7.488E-03	6.207E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
4	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
5	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
6	2.512E-03	7.488E-03	6.207E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
7	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
8	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02

TRANSIENT BEHAVIOR RESULTS MIXER CONCENTRATION GIVEN

TIME = .00 MINUTES

AQUEOUS PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	TEMPERATURE (CENTIGRADE)
1	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
2	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
3	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
E 3	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00			
4	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
5	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
6	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
E 6	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00			
7	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
8	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01

ORGANIC PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	INVENTORY CHANGE %)
1	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
2	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
3	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
4	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
5	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
6	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
7	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
8	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02

TIME = 302.00 MINUTES

AQUEOUS PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	TEMPERATURE (CENTIGRADE)
1	4.862E-01	1.275E+01	7.828E-01	2.020E-03	6.511E-02	4.980E-01	1.126E+00	6.024E-03	2.500E+01
2	4.857E-01	1.485E+01	1.070E+00	1.349E-01	3.039E-01	4.976E-01	1.130E+00	6.030E-03	2.500E+01
3	4.847E-01	1.488E+01	1.172E+00	6.166E-01	1.377E+00	4.972E-01	1.131E+00	6.033E-03	2.500E+01
E 3	4.385E-01	8.790E+00	2.450E-05	1.799E+00	7.453E+00	4.971E-01			
4	4.616E-01	1.393E+01	1.237E+00	2.043E+00	5.656E+00	4.960E-01	1.131E+00	6.040E-03	2.500E+01
5	4.958E-01	1.399E+01	1.589E+00	3.783E+00	4.807E+00	4.950E-01	1.136E+00	6.052E-03	2.500E+01
6	5.453E-01	1.284E+01	1.929E+00	7.086E+00	4.265E+00	4.935E-01	1.142E+00	6.071E-03	2.500E+01
E 6	5.118E-01	7.884E+00	4.245E-05	9.050E+00	9.211E+00	4.932E-01			
7	5.934E-01	1.116E+01	2.431E+00	1.027E+01	6.494E+00	4.910E-01	1.147E+00	6.094E-03	2.500E+01
8	7.623E-01	1.010E+01	2.833E+00	1.160E+01	3.885E+00	4.885E-01	1.154E+00	6.126E-03	2.500E+01

ORGANIC PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	INVENTORY CHANGE %)
1	6.278E-02	5.401E+01	9.666E-01	2.918E-05	6.417E-03	.000E+00	8.941E-01	1.787E-02	9.507E-02
2	5.872E-02	5.823E+01	1.226E+00	1.781E-03	2.780E-02	.000E+00	9.003E-01	1.789E-02	6.490E-02
3	5.836E-02	5.890E+01	1.358E+00	8.139E-03	1.274E-01	.000E+00	9.015E-01	1.790E-02	4.625E-02
4	5.689E-02	5.883E+01	1.534E+00	2.822E-02	5.603E-01	.000E+00	9.017E-01	1.790E-02	3.000E-02
5	6.028E-02	6.000E+01	2.008E+00	5.178E-02	4.858E-01	.000E+00	9.043E-01	1.792E-02	2.999E-02
6	6.754E-02	5.973E+01	2.663E+00	1.012E-01	4.711E-01	.000E+00	9.053E-01	1.793E-02	2.325E-02
7	7.585E-02	5.878E+01	3.838E+00	1.565E-01	8.215E-01	.000E+00	9.063E-01	1.794E-02	1.842E-02
8	9.706E-02	5.917E+01	5.059E+00	1.785E-01	5.569E-01	.000E+00	9.097E-01	1.797E-02	1.738E-02

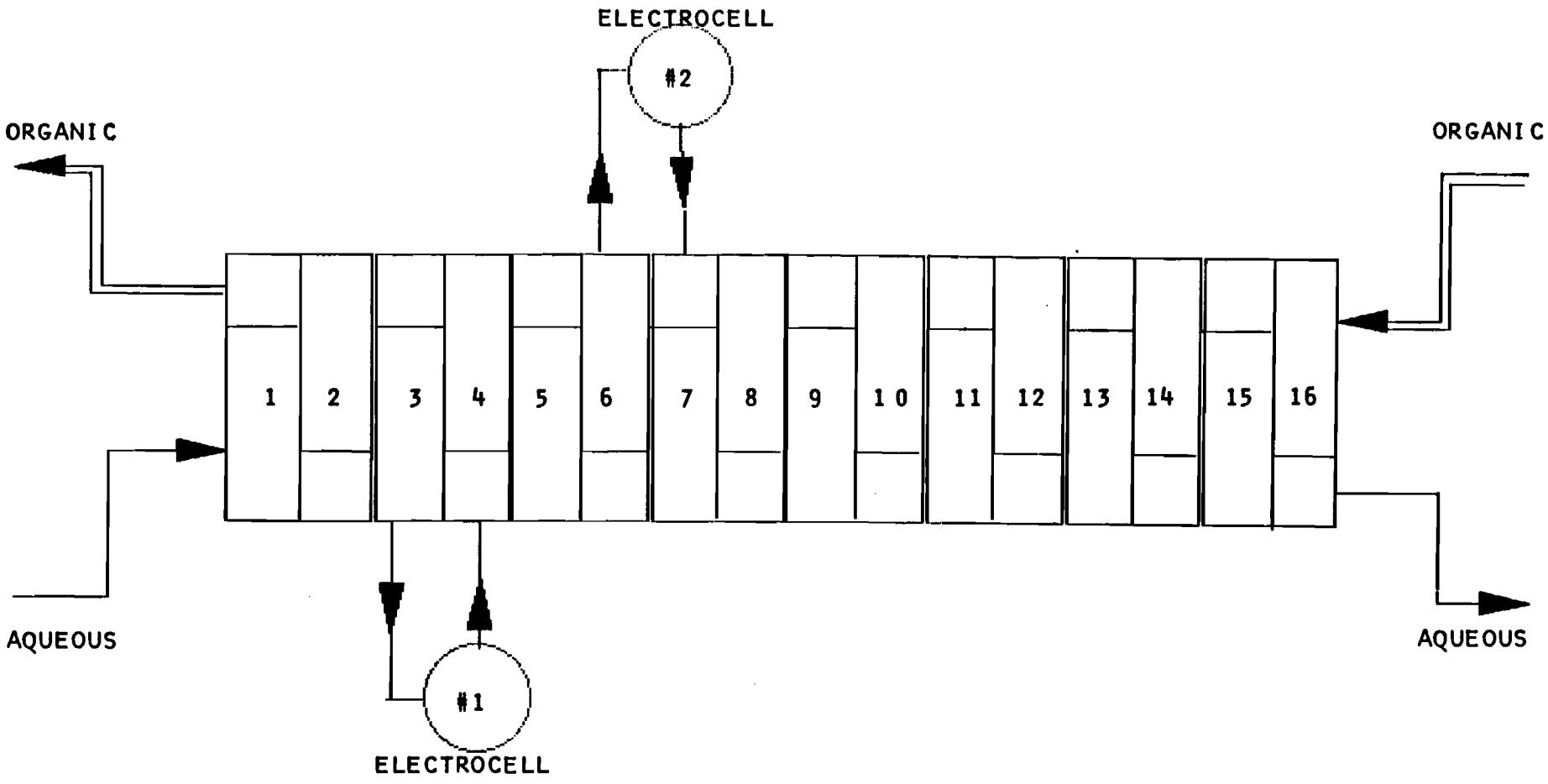

Figure C.3 Sixteen-Stage U-Pu Partitioning Cascade

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 TEST SAMPLE No. 3

  INPUT FILE "SAMPLE3.INP"

 216 0.3    25.0     0 0 0 6
 3 6 99   0.047 0.075   0.075    0.50     0.00     0.50
THIS IS A TEST EXAMPLE OF SEPHIS5-GT
CASE 3 - SIXTEEN-STAGE CONTACTOR WITH TWO ELECTROCELLS
1.0      550.0    550.0 0.1      1 0 2 2 0 1 0
 1 1  0.006 0.5      000.0  0.00    0.0     0.0     0.5     25.0     1
16 0  0.018 0.15      58.4  5.84    0.0     0.0     0.0     25.0     0
 1 1 0.01            1
 2 1 0.06            0
 0.0    0.0

OUTPUT FILE "SAMPLE3.OUT"

 CALCULATIONS FOR A SOLVENT EXTRACTION PROCESS HAVING INTERACTING SOLUTES


 PUREX PROCESS
 THIS IS A TEST EXAMPLE OF SEPHIS5-GT
 CASE 3 - SIXTEEN-STAGE CONTACTOR WITH TWO ELECTROCELLS
-DTHETA =    1.000 MINUTES PER TIME INCREMENT
 DPRINT =  550.000 MINUTES BETWEEN PRINTING OF PROFILES
 IFAST = 1  THE FAST INTEGRATION TECHNIQUE WILL BE USED
 THIS TIME PERIOD WILL END WHEN TIME = TSTOP =  550.000 MINUTES, OR A TOLERANCE OF TOL =   .1000 % PER MINUTE IS REACHED
 NUMBER OF STAGES =  16
 NEWIN = 1 NEW INPUT FLOWS WILL BE GIVEN
 NEWOUT = 0  OUTPUT STREAMS WILL BE UNCHANGED
 IVOLM = 2 TOTAL MIXER VOLUME GIVEN
 IVOLS = 2 TOTAL SETTLER VOLUME GIVEN
 IPRO = 0  A NEW INITIAL PROFILE WILL NOT BE READ
 IPNCH = 0   NO PUNCHED CARD OUTPUT
 NSTR = 0   NO UNUSUAL ROUTING PATTERN
 TEMPI =  2.500E+01  INITIAL & DEFAULT TEMPERATURE
 IRXN = 6  ELECTROCELL PLUS REDUCTION BY U(IV)
 ELECTROCELL BETWEEN STAGE  3 AND STAGE  4
 ELECTROCELL BETWEEN STAGE  6 AND STAGE  7
 VC   =   .0470 LITER -- VOLUME OF ELECTROCELL
 BRU  =   .0750       -- ELECTRICAL FACTOR FOR U(VI)
 BRPU =   .0750       -- ELECTRICAL FACTOR FOR PU(IV)
 OC   =   .5000       -- GEOMETRIC FACTOR OF CATHODE
 OA   =   .0000       -- GEOMETRIC FACTOR OF ANODE
 GAM  =   .5000       -- REACTION RATE CONSTANT FOR U(IV)+ PU(IV) REACTION

-FEED & PRODUCT      STAGE  NITRIC ACID     U (VI)        PU (IV)       PU (III)       U (IV)     NITRATE ION    FLOW RAT     TEMP
 STREAM DATA          NO.     (MOL/L)      (G/L)         (G/L)         (G/L)         (G/L)        (MOL/L)       (L/MIN)       (C)
            AQUEOUS   1     5.000E-01     .000E+00      .000E+00      .000E+00      .000E+00     5.000E-01     6.000E-03      25.0
          30.0 % TBP  16    1.500E-01    5.840E+01     5.840E+00      .000E+00      .000E+00      .000E+00     1.800E-02      25.0
```

STAGE NO.	MIXER VOLUME BY PHASE		SETTLER VOLUME BY PHASE		MIXER FLOW RATE		INTERSTAGE FLOW RATE	
	AQUEOUS	ORGANIC	AQUEOUS	ORGANIC	AQUEOUS	ORGANIC	AQUEOUS	ORGANIC
1	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
2	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
3	2.512E-03	7.488E-03	6.207E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
4	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
5	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
6	2.512E-03	7.488E-03	6.207E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
7	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
8	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
9	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
10	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
11	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
12	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
13	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
14	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
15	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02
16	2.512E-03	7.488E-03	1.507E-02	4.493E-02	5.814E-03	1.733E-02	5.814E-03	1.733E-02

TRANSIENT BEHAVIOR RESULTS MIXER CONCENTRATION GIVEN

TIME = .00 MINUTES

AQUEOUS PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	TEMPERATURE (CENTIGRADE)
1	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
2	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
3	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
E 3	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00			
4	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
5	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
6	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
E 6	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00			
7	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
8	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
9	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
10	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
11	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
12	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
13	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
14	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
15	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01
16	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	9.971E-01	5.814E-03	2.500E+01

ORGANIC PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	INVENTORY CHANGE %)
1	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
2	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
3	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
4	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
5	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
6	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
7	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
8	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
9	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
10	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
11	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
12	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
13	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
14	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
15	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02
16	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	.000E+00	8.171E-01	1.749E-02	2.000E+02

- TIME = 463.00 MINUTES

AQUEOUS PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	TEMPERATURE (CENTIGRADE)
1	4.847E-01	1.285E+01	7.386E-02	.000E+00	9.492E-02	4.982E-01	1.125E+00	6.021E-03	2.500E+01
2	4.792E-01	1.501E+01	9.714E-02	9.127E-02	4.265E-01	4.979E-01	1.128E+00	6.025E-03	2.500E+01
3	4.657E-01	1.524E+01	1.298E-01	2.067E-01	1.818E+00	4.978E-01	1.128E+00	6.026E-03	2.500E+01
E 3	4.113E-01	8.694E+00	1.996E-06	3.379E-01	8.361E+00	4.978E-01			
4	4.189E-01	1.471E+01	1.654E-01	3.590E-01	6.921E+00	4.971E-01	1.126E+00	6.027E-03	2.500E+01
5	4.167E-01	1.547E+01	2.218E-01	5.540E-01	7.226E+00	4.969E-01	1.128E+00	6.029E-03	2.500E+01
6	4.057E-01	1.538E+01	2.890E-01	8.160E-01	8.601E+00	4.968E-01	1.128E+00	6.030E-03	2.500E+01
E 6	3.519E-01	8.826E+00	5.122E-07	1.110E+00	1.515E+01	4.968E-01			
7	3.632E-01	1.476E+01	3.642E-01	1.156E+00	1.346E+01	4.960E-01	1.126E+00	6.031E-03	2.500E+01
8	3.693E-01	1.549E+01	4.852E-01	1.586E+00	1.317E+01	4.957E-01	1.128E+00	6.035E-03	2.500E+01
9	3.783E-01	1.545E+01	6.310E-01	2.159E+00	1.282E+01	4.954E-01	1.130E+00	6.039E-03	2.500E+01
10	3.905E-01	1.520E+01	8.127E-01	2.907E+00	1.235E+01	4.951E-01	1.131E+00	6.043E-03	2.500E+01
11	4.070E-01	1.485E+01	1.037E+00	3.877E+00	1.174E+01	4.946E-01	1.133E+00	6.048E-03	2.500E+01
12	4.301E-01	1.438E+01	1.309E+00	5.128E+00	1.092E+01	4.941E-01	1.136E+00	6.055E-03	2.500E+01
13	4.645E-01	1.375E+01	1.625E+00	6.744E+00	9.771E+00	4.933E-01	1.139E+00	6.065E-03	2.500E+01
14	5.202E-01	1.288E+01	1.965E+00	8.909E+00	8.072E+00	4.921E-01	1.144E+00	6.079E-03	2.500E+01
15	6.169E-01	1.148E+01	2.247E+00	1.298E+01	5.358E+00	4.900E-01	1.152E+00	6.105E-03	2.500E+01
16	7.781E-01	9.847E+00	2.727E+00	1.419E+01	3.049E+00	4.876E-01	1.158E+00	6.135E-03	2.500E+01

ORGANIC PHASE

STAGE NO.	NITRIC ACID (MOL/L)	U (VI) (G/L)	PU (IV) (G/L)	PU (III) (G/L)	U (IV) (G/L)	NITRATE ION (MOL/L)	DENSITY (G/ML)	MIXER FLOW (L/MIN)	INVENTORY CHANGE %)
1	6.280E-02	5.439E+01	9.111E-02	.000E+00	9.344E-03	.000E+00	8.932E-01	1.786E-02	1.846E-02
2	5.821E-02	5.865E+01	1.108E-01	1.209E-03	3.883E-02	.000E+00	8.990E-01	1.788E-02	4.163E-02
3	5.621E-02	5.935E+01	1.477E-01	2.718E-03	1.652E-01	.000E+00	9.000E-01	1.789E-02	4.947E-02
4	5.138E-02	5.941E+01	1.953E-01	4.858E-03	6.525E-01	.000E+00	9.000E-01	1.789E-02	9.754E-02
5	4.995E-02	6.091E+01	2.556E-01	7.269E-03	6.649E-01	.000E+00	9.022E-01	1.790E-02	6.329E-02
6	4.872E-02	6.113E+01	3.365E-01	1.076E-02	7.998E-01	.000E+00	9.026E-01	1.790E-02	5.746E-02
7	4.437E-02	6.106E+01	4.421E-01	1.572E-02	1.305E+00	.000E+00	9.026E-01	1.790E-02	9.921E-02
8	4.405E-02	6.252E+01	5.753E-01	2.089E-02	1.248E+00	.000E+00	9.048E-01	1.791E-02	6.242E-02
9	4.502E-02	6.269E+01	7.530E-01	2.842E-02	1.222E+00	.000E+00	9.054E-01	1.791E-02	5.343E-02
10	4.660E-02	6.258E+01	9.849E-01	3.855E-02	1.196E+00	.000E+00	9.057E-01	1.792E-02	4.557E-02
11	4.879E-02	6.237E+01	1.285E+00	5.196E-02	1.162E+00	.000E+00	9.060E-01	1.792E-02	3.849E-02
12	5.188E-02	6.209E+01	1.670E+00	6.973E-02	1.114E+00	.000E+00	9.064E-01	1.792E-02	3.201E-02
13	5.649E-02	6.172E+01	2.162E+00	9.350E-02	1.040E+00	.000E+00	9.069E-01	1.793E-02	2.577E-02
14	6.402E-02	6.123E+01	2.784E+00	1.269E-01	9.154E-01	.000E+00	9.076E-01	1.794E-02	1.877E-02
15	7.759E-02	6.057E+01	3.569E+00	1.937E-01	6.820E-01	.000E+00	9.085E-01	1.795E-02	8.017E-02
16	9.972E-02	5.937E+01	5.025E+00	2.209E-01	4.513E-01	.000E+00	9.100E-01	1.797E-02	3.969E-02